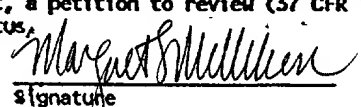


Form PTO-1390		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER US 17017
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		U.S. APPLICATION NO. 09/600363	
INTERNATIONAL APPLICATION NO. PCT/AU99/00036	INTERNATIONAL FILING DATE January 19, 1999	PRIORITY DATE CLAIMED January 19, 1998	
TITLE OF INVENTION PROCESS FOR INCREASING THE MELT STRENGTH OF POLYPROPYLENE		DATE: July 14, 2000	
APPLICANT(S) FOR DO/EO/US Polymers Australia Pty Limited			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<ol style="list-style-type: none"> <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. <input type="checkbox"/> This a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. <input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1). <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) <ol style="list-style-type: none"> <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). <input checked="" type="checkbox"/> has been transmitted by the International Bureau. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). <input type="checkbox"/> have been transmitted by the International Bureau. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. <input type="checkbox"/> have not been made and will not be made. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). 			
Items 11. to 16. below concern other document(s) or information included:			
11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.			
12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.			
13. <input type="checkbox"/> A FIRST preliminary amendment. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.			
14. <input type="checkbox"/> A substitute specification.			
15. <input type="checkbox"/> A change of power of attorney and/or address letter.			
16. <input checked="" type="checkbox"/> Other items or information: See enclosed cover letter identifying inventors.			

U.S. Application No. 09/600363		International Application No. PCT/AU99/00036		Attorney's Docket No. US 17017	
17. <input checked="" type="checkbox"/> The following fees are submitted: Basic National Fee (37 CFR 1.492(a)(1)-(5)): Search Report has been prepared by the EPO or JPO. \$ 840 International preliminary examination fee paid to USPTO (37 CFR 1.482). 670 No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) 760 Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO 970 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4). 96 <div style="text-align: right;">ENTER APPROPRIATE BASIC FEE AMOUNT =</div>				CALCULATIONS	PTO USE ONLY
Surcharge of \$130.00 for furnishing the oath or declaration later than [] 20 [] 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$ -0-	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	19 - 20	-0-	x \$18	\$ -0-	
Indep. claims	2 - 3	-0-	x \$78	\$ -0-	
Multiple dependent claim(s) (if applicable)			+ \$260	\$ 260	
TOTAL OF ABOVE CALCULATIONS =				\$ 1100	
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).				\$	
SUB TOTAL =				\$ 1100	
Processing fee \$130.00 for furnishing the English translation later than [] 20 [] 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
TOTAL NATIONAL FEE =				\$ 1100	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				\$	
TOTAL FEES ENCLOSED =				\$ 1100	
				Amount to be: refunded	\$
				charged	\$
a. <input type="checkbox"/> A check in the amount of \$_____ to cover the above fees is enclosed. b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. <u>08-2336</u> in the amount of \$ <u>1100</u> to cover the above fees. Two copies of this sheet are enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>08-2336</u> . Two copies of this sheet are enclosed.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to review (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO: Montell North America Inc. Intellectual Property & Licensing 912 Appleton Road Elkton, Maryland 21921 phone: 410-996-1646 fax: 410-996-1560				<div style="text-align: center;">  Signature Margaret S. Millikin Name 38,969 Registration Number July 14, 2000 Date </div>	

- 1 -

PROCESS

FOR INCREASING THE MELT STRENGTH OF POLYPROPYLENE

5 The present invention relates to polypropylene homopolymers and copolymers. In particular, the present invention relates to a process for increasing the melt strength and/or the extensional melt viscosity of said polymers by melt phase processing.

The melt strength and extensional viscosity of linear or straight chain polymers, such as polypropylene, decreases rapidly with temperature. By contrast, polymers such as low density polyethylene which are highly branched retain relatively high melt strengths and extensional viscosities. It is generally understood that the difference in melt strengths and extensional viscosities is attributable to the presence of long chain branching in polymers such as low density polyethylene. Long chain branching allows a greater degree of chain entanglement.

A number of methods for increasing the melt strength/extensional viscosity of polypropylene and related polymers through the introduction of branching or a limited degree of crosslinking in a process involving reactive extrusion have been proposed and are summarised in a recent paper by Wang et al. (Wang, X., Tzoganakis, C., and Rempel, G.L., *J. Appl. Polym. Sci.*, 1996, 61, 1395). One such process involves the reactive extrusion of polypropylene with a polyfunctional monomer/initiator combination. For example, the use of pentaerythritol triacrylate in combination with 2,5-dimethyl-2,5-di(t-butylperoxy)hexane (DHBP).

25 White (US 5578682) has disclosed the use of various polyunsaturated crosslinking agents (for example, bismaleimide derivatives) in combination with free radical initiators to achieve an increase in the melt strength various polymers.

It is well known that the melt phase processing of polypropylene leads to mechanochemical degradation. The processing of polypropylene in the presence of free radical initiators

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provides an increased rate of degradation. This controlled degradation of polypropylene is used commercially for the production of controlled rheology resins having reduced polydispersity and reduced die swell (Lambla, M. in *Comprehensive Polymer Science*, Pergamon, New York 1992, vol Suppl. 1, p 619; Hogt, A.H., Meijer, J., Jelinic, J. in
5 Reactive Modifiers for Polymers, Al-Malaika, S. Ed., Chapman & Hall, London, 1996, p 84.). The degradation of polypropylene as described therein results in a lowering of melt strength.

The batch modification of polypropylene to produce crosslinked (insoluble) polypropylene by
10 treatment with peroxides is described by Borsig et al. (Borsig, E., Fiedlerova, A., Lazar, M. J., *Macromol. Sci, Chem.*, 1981, A16, 513). Initiators which produce benzoyloxy radicals or phenyl radicals are described as being more efficient in inducing crosslinking or grafting than those which produce t-butoxy or alkyl radicals. The process requires the use of high levels of peroxide. The use of polyfunctional monomers as coagents to retard degradation and
15 enhance crosslinking is described by Chodak, I.; Fabianova, K.; Borsig, E.; Lazar, M. Agnew. Makromol. Chem., 1978, 69, 107.

DeNicola (EP 384331A2) has disclosed a means to produce a branched propylene polymer material showing a nett increase in the weight average molecular weight by solid state
20 modification of predominantly isotactic semi-crystalline linear polypropylene. The process described in EP384331A2 involves blending peroxides with short half lives (eg peroxy dicarbonates) with linear propylene polymer in a mixing vessel at temperatures from 23°C to 120°C in an inert atmosphere and continuing to mix for a period of time until the peroxide decomposes and polymer fragmentation and branching occurs without significant gelation of
25 the polymer. DeNicola states that at temperatures greater than 120°C no branching or melt strength enhancement is achieved.

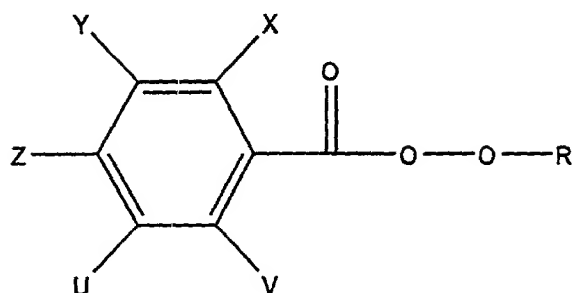
U.S. 5,464,907 teaches that certain unsaturated maleate or itaconate derived peroxides may be used to induce grafting in polypropylene and α -olefin copolymers. They report that use

- 3 -

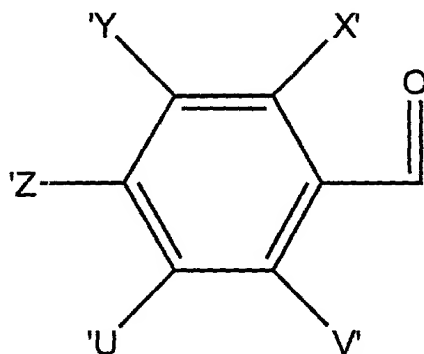
of other peroxides generally results in chain degradation.

- Polypropylene is also known to undergo substantial degradation during melt phase grafting of monofunctional monomers, for example maleic anhydride and glycidyl methacrylate. It has also been reported that the degradation that accompanies grafting of these monomers to polypropylene may be reduced by the addition of relatively high concentrations of certain comonomers including styrene (see, for example Sun, Y.-J., Hu, G.-H., and Lambla, M., *Angew. Makromol. Chem.*, 1995, 229, 1; Chen, L-F., Wong, B. and Baker, W.E. *Polym. Eng. Sci.* 1996, 36, 1594.) Sun et al. report that there is degradation (as indicated by an overall decrease in molecular weight) when styrene alone is grafted onto polypropylene even when a relatively high concentration is used (4 moles/100g PP). Either 2,5-dimethyl-2,5-(t-butylperoxy)hex-3-yne or 2,5-dimethyl-2,5(t-butylperoxy-hexane(DHBP) was used as the initiator in these experiments.
- We have found that melt mixing polypropylene homopolymer or ethylene-polypropylene copolymer in the presence of a suitable initiator provides one or more of the following: increased melt strength; increased extensional viscosity; increased molecular weight; and broadened molecular weight distribution.
- According to the present invention there is provided a process for modifying a polypropylene (co)polymer wherein said process comprises melt mixing the polypropylene (co)polymer in the presence of an initiator wherein said initiator is selected from the group defined by formula 1:

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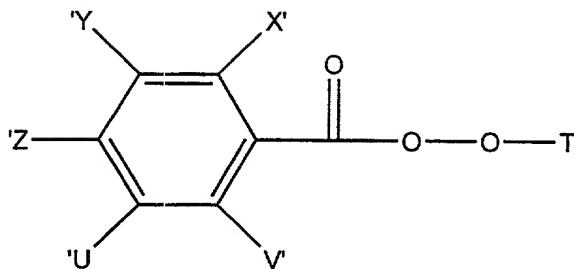
**Formula 1**

wherein R is selected from the group consisting of optionally substituted C₁ to C₁₈ acyl,
5 optionally substituted C₁ to C₁₈ alkyl, aryl defined by formula 2,

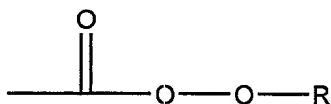
**Formula 2**

10 and groups of formula 3,

- 5 -

**Formula 3**

wherein U, V, X, Y, Z, U', V', X', Y' and Z' are independently selected from the group
 5 consisting hydrogen, halogen, C1-C18 alkyl, C1-C18 alkoxy, aryloxy, acyl, acyloxy, aryl,
 carboxy, alkoxycarbonyl, aryloxycarbonyl, trialkyl silyl, hydroxy,
 or a moiety of formula 4,

**Formula 4**

and wherein T is alkylene.

Advantageously the thus formed modified polypropylene may be obtained without the
 associated production of significant and detrimental amounts of gels.

15

Polymers suitable for use in the present invention include a wide variety of polypropylene
 homopolymers, copolymers and blends containing one or more polypropylene homopolymers
 and/or copolymers.

20 Suitable polypropylene homopolymers include isotactic polypropylene, atactic polypropylene
 and syndiotactic polypropylene. Commercial isotactic polypropylene having a proportion of
 meso/dyads of greater than 90% is preferably used in the process of the present invention.

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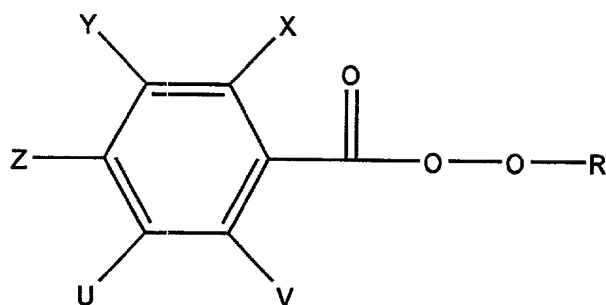
meso/dyads of greater than 90% is preferably used in the process of the present invention. Isotactic polypropylene is a semi-crystalline polymer having a number of properties which have made it one of the most widely used commercial polymers. These properties include heat resistance, stress cracking resistance, chemical resistance, toughness, and low manufacturing costs. However, the melt strength of isotactic polypropylene as measured directly by extensional viscosity or use of a commercial melt strength tester or indirectly by more qualitative measures such as drop time or die swell ratio is relatively low. This relatively low melt strength limits the use of polypropylene in applications such as foam extrusion, thermoforming and film blowing. In order to use polypropylene in such applications it is necessary to employ sophisticated processing equipment. The present invention now permits this already widely used commercial polymer to be used in an even wider range of applications.

Polypropylene copolymers include copolymers of propylene and other monomers with such other monomers being present preferably in amounts of up to 10%wt/wt. A preferred comonomer is ethylene.

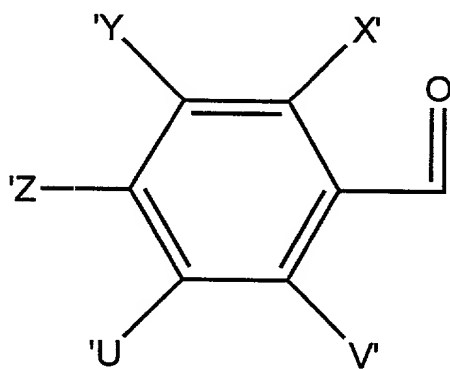
The present invention is also applicable to other polymers comprising α -olefin monomers. It is preferable that any such α -olefins are present in the polymer to be modified in amounts in excess of 90%wt/wt. α -olefins include propene, 1-butene, 1-pentene and 1-hexene.

The initiators for use in the present invention may be selected from the group defined by formula 1.

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**Formula 1**

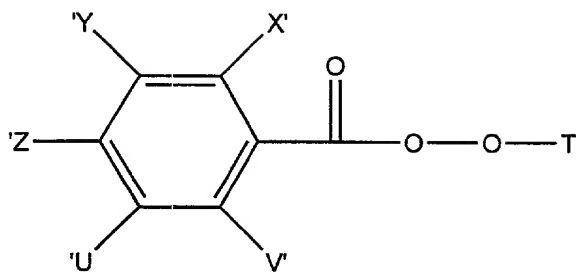
5 wherein R is selected from the group consisting of optionally substituted C₁ to C₈ acyl, optionally substituted C₁ to C₁₈ alkyl, aroyl defined by formula 2,

**Formula 2**

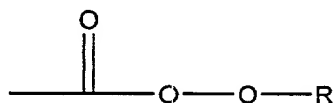
10

and groups of formula 3,

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**Formula 3**

wherein U, V, X, Y, Z, U', V', X', Y' and Z' are independently selected from the group
 5 consisting hydrogen, halogen, C1-C18 alkyl, C1-C18 alkoxy, aryloxy, acyl, acyloxy, aryl,
 carboxy, alkoxycarbonyl, aryloxycarbonyl, trialkyl silyl, hydroxy,
 or a moiety of formula 4,



10

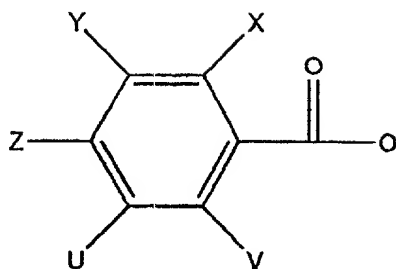
Formula 4

and wherein T is alkylene.

The alkyl, including acyl and alkoxy, groups included in the initiators of formula 1 may
 15 include hetero atoms within the carbon chain (eg polyalkylene oxide) and may be branched
 or unbranched and may be substituted with one or more groups such as with alkyl, aryl,
 alkoxy or halogen substituents.

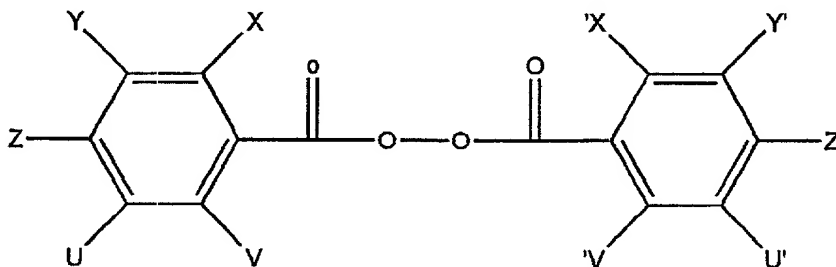
Without wishing to be bound by theory, it is believed that the aryloxy radical of formula 5
 20

- 9 -

**Formula 5**

,where U, V, X, Y and Z are as hereinabove defined, provide the surprising increase in melt strength. Other compounds which generate these aryloxy radicals may also be used in the present invention.

A preferred class of initiators of formula 1 are diaryl peroxides of formula 6.



10

Formula 6

where X, Y, Z, U, V, X', Y', Z', U', V' are independently selected from the group consisting of hydrogen and C₁ - C₁₈ alkyl where at least one of X, Y, Z, U, V and X', Y', Z', U', V' are not hydrogen.

15

Diaryl peroxides of formula 6 include Dibenzoyl peroxide, o,o'-Bis(methylbenzoyl) peroxide, p,p'-Bis(methylbenzoyl) peroxide, M,M'-Bis(methylbenzoyl) peroxide, o,m'-Bis(methylbenzoyl)

- peroxide, o,p'-Bis(methylbenzoyl) peroxide, m,p'-Bis(methylbenzoyl) peroxide, Bis(ethylbenzoyl) peroxide (all isomers), Bis(propylbenzoyl) peroxide (all isomers), Bis(butylbenzoyl) peroxide (all isomers), Bis(pentylbenzoyl) peroxide (all isomers), Bis(hexylbenzoyl) peroxide (all isomers), Bis(heptylbenzoyl) peroxide (all isomers), Bis(octylbenzoyl) peroxide (all isomers),
- 5 Bis(nonylbenzoyl) peroxide (all isomers), Bis(methoxybenzoyl) peroxide (all isomers), Bis(ethoxybenzoyl) peroxide (all isomers), Bis(propoxybenzoyl) peroxide (all isomers), Bis(butoxybenzoyl) peroxide (all isomers), Bis(pentoxymethylbenzoyl) peroxide (all isomers), Bis(hexyloxybenzoyl) peroxide (all isomers), Bis(heptyloxybenzoyl) peroxide (all isomers), Bis(octyloxybenzoyl) peroxide (all isomers), Bis(nonyloxybenzoyl) peroxide (all isomers),
- 10 Bis(chlorobenzoyl) peroxide (all isomers), Bis(fluorobenzoyl) peroxide (all isomers), Bis(bromobenzoyl) peroxide (all isomers), Bis(dimethylbenzoyl) peroxide (all isomers), Bis(trimethylbenzoyl) peroxide (all isomers), Bis(tert-butylbenzoyl)peroxide (all isomers), Bis(di-tert-butylbenzoyl)peroxide (all isomers), Bis(tert-butoxybenzoyl)peroxide (all isomers), Bis(ditrimethylsilylbenzoyl) peroxide (all isomers), Bis(heptafluoropropylbenzoyl) peroxide (all isomers), Bis(2,6-dimethyl-4- trimethylsilyl benzoyl) peroxide and isomers, 2,2'(dioxydicarbonyl)
- 15 bis - Benzoic acid dibutyl ester where the term "all isomers" refers to any variation in the position of the ring substituent as well as the structure of the substituent itself i.e. for propyl; n-propyl and isopropyl.
- 20 Examples of aromatic peresters of formula 1 include the following: tert-butyl perbenzoate, tert-butyl (methyl)perbenzoate (all isomers), tert-butyl (ethyl)perbenzoate (all isomers), tert-butyl (octyl)perbenzoate (all isomers), tert-butyl (nonyl)perbenzoate (all isomers), tert-amyl perbenzoate, tert-amyl (methyl)perbenzoate (all isomers), tert-amyl (ethyl)perbenzoate (all isomers), tert-amyl (octyl)perbenzoate (all isomers), tert-amyl (nonyl)perbenzoate (all isomers),
- 25 tert-amyl (methoxy)perbenzoate (all isomers), tert-amyl (octyloxy)perbenzoate (all isomers), tert-amyl (nonyloxy)perbenzoate (all isomers), 2-ethylhexyl perbenzoate, 2-ethylhexyl (methyl)perbenzoate (all isomers), , 2-ethylhexyl (ethyl)perbenzoate (all isomers), 2-ethylhexyl (octyl)perbenzoate (all isomers), , 2-ethylhexyl (nonyl)perbenzoate (all isomers), 2-ethylhexyl (methoxy)perbenzoate (all isomers), 2-ethylhexyl (ethoxy)perbenzoate (all isomers), 2-
- 30 ethylhexyl (octyloxy)perbenzoate (all isomers), 2-ethylhexyl (nonyloxy)perbenzoate (all isomers)

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The initiators for use in the present invention also include compounds of formula 1 where at least one of U, V, X, Y, Z, U', V', X' Y' and Z' is a moiety of formula 4 where R is as defined above. Preferably there is no more than one moiety of formula 4 per aromatic ring. Such initiators are di or higher functional peroxides and may include polymeric peroxides
5 such as Bis (tertbutylmonoperoxy phthaloyl) diperoxy terephthalate, Bis (tertamylmonoperoxy phthaloyl) diperoxy terephthalate diacetyl phthaloyl diperoxide, dibenzoyl phthaloyl diperoxide, bis(4 methylbenzoyl) phthaloyl diperoxide, diacetyl terephthaloyl di peroxide, dibenzoyl terephthaloyl diperoxide, Poly[dioxycarbonyldioxy(1,1,4,4-tetramethyl-1,4-butanediyl)] peroxide.

10

It is described that the initiators are selected such that it has an appropriate decomposition temperature (half life), solubility, and reactivity and such that the groups R, T, X, Y, Z, U, V, X', Y', Z', U', V' give no adverse reaction under the conditions of the process. Preferred
15 peroxides will have a 0.1 hour half life in the range 100 - 170°C.

The amount of initiator used in the process of the present invention should be an effective amount to achieve the desired increase in melt strength. Melt strength is considered in the art to be an indication of long-chain branching in polyolefins. It is preferable in the process
20 of the present invention that long-chain branching predominates over crosslinking in the reaction between the initiator and the polypropylene (co)polymer. Crosslinking of the polypropylene (co)polymer may result in the formation of gels which disrupt the appearance of the polypropylene (co)polymer. In the process of the present invention it is desirable to control the degree and distribution of crosslinking and keep the level of crosslinking as
25 uniform and as low as necessary to produce the desired effects. The amount of crosslinking which occurs in the polypropylene (co)polymer is dependant upon the amount of initiator melt mixed with the polypropylene (co)polymer. The amount of crosslinking is also dependent upon the degree of mixing as any regions high in initiator concentration will result in excessive localised crosslinking and the formation of gels. It is desirable that good
30 distributive and dispersive mixing be employed to promote even distribution of the initiator

- 12 -

in the polypropylene (co)polymer so as to minimise the variation in initiator concentration throughout the polypropylene (co)polymer and reduce the likelihood of the formation of gels.

Preferably the initiator will be present in the range of from 0.004 to 0.25 moles of initiator per kg of the polypropylene homopolymer or copolymer (polypropylene (co)polymer). The more preferred range being from 0.006 to 0.10 moles of initiator per kg. of the polypropylene (co)polymer and even more preferred range being from 0.01 to 0.05 moles of initiator per kg of the polypropylene (co)polymer.

- 10 The initiator is preferably introduced into the polymer melt directly, either neat (as a powder or a liquid), dispersed or dissolved in a suitable medium (for example, dissolved in 2-butanone) or adsorbed on polymer pellets or powder which are added as a masterbatch. It is desirable that the initiator is rapidly mixed with the polymer melt at a rate in keeping with the half life of the initiator at the processing temperature of the polypropylene (co)polymer.

15

The initiator may be added either alone, or along with the polypropylene (co)polymer, or with any other polymer, additive or filler, so that the polymer melts and mixes with the initiator as it is decomposing. When the initiator is fed to the main feed throat of the extruder it is preferred to have a barrel temperature which is relatively low in the region adjacent to the main feed throat and increases towards the die to prevent premature decomposition of the peroxide.

Preferably the initiators for use in the present invention are selected from the group consisting of Dibenzoyl peroxide, o,o'-Bis(methylbenzoyl) peroxide, p,p'-Bis(methylbenzoyl) peroxide, o,o'-Bis(methylbenzoyl) peroxide, o,m'-Bis(methylbenzoyl) peroxide, o,p'-Bis(methylbenzoyl) peroxide, m,p'-Bis(methylbenzoyl) peroxide, Bis(ethylbenzoyl) peroxide (all isomers), Bis(propylbenzoyl) peroxide (all isomers), Bis(butylbenzoyl) peroxide (all isomers), Bis(pentylbenzoyl) peroxide (all isomers), Bis(hexylbenzoyl) peroxide (all isomers), Bis(heptylbenzoyl) peroxide (all isomers), Bis(octylbenzoyl) peroxide (all isomers), Bis(nonylbenzoyl) peroxide (all isomers), Bis(methoxybenzoyl) peroxide (all isomers),

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Bis(ethoxybenzoyl) peroxide (all isomers), Bis(propoxybenzoyl) peroxide (all isomers), Bis(butoxybenzoyl) peroxide (all isomers), Bis(pentoxybenzoyl) peroxide (all isomers), Bis(hexyloxybenzoyl) peroxide (all isomers), Bis(heptyloxybenzoyl) peroxide (all isomers), Bis(octyloxybenzoyl) peroxide (all isomers), Bis(nonyloxybenzoyl) peroxide (all isomers),

5 Bis(chlorobenzoyl) peroxide (all isomers), Bis(fluorobenzoyl) peroxide (all isomers), Bis(bromobenzoyl) peroxide (all isomers), Bis(dimethylbenzoyl) peroxide (all isomers), Bis(trimethylbenzoyl) peroxide (all isomers), Bis(tert-butylbenzoyl)peroxide (all isomers), Bis(di-tert-butylbenzoyl)peroxide (all isomers), Bis(tertbutoxybenzoyl)peroxide (all isomers), Bis(ditrimethylsilylbenzoyl) peroxide (all isomers), Bis(heptafluoropropylbenzoyl) peroxide (all

10 isomers), Bis(2,4-dimethyl-6- trimethylsilyl benzoyl) peroxide and isomers tert-amyl perbenzoate, tert-amyl (methyl)perbenzoate (all isomers), tert-amyl (ethyl)perbenzoate (all isomers), tert-amyl (octyl)perbenzoate (all isomers), tert-amyl (nonyl)perbenzoate (all isomers), tert-amyl (methoxy)perbenzoate (all isomers), tert-amyl (octyloxy)perbenzoate (all isomers), tert-amyl (nonyloxy)perbenzoate (all isomers), Bis (tertamylmonoperoxy phthaloyl) diperoxy terephthalate,

15 diacetyl phthaloyl diperoxide, dibenzoyl phthaloyl diperoxide, bis(4-methylbenzoyl) phthaloyl diperoxide, diacetyl terephthaloyl di peroxide and dibenzoyl terephthaloyl diperoxide.

More preferably the initiators are selected from the group consisting of dibenzoyl peroxide, o,o'-Bis(methylbenzoyl) peroxide, p,p'-Bis(methylbenzoyl) peroxide, M,M'-Bis(methylbenzoyl)

20 peroxide, o,m'-Bis(methylbenzoyl) peroxide, o,p'-Bis(methylbenzoyl) peroxide, m,p'-Bis(methylbenzoyl) peroxide.

The initiators may optionally be used in combination with one or more monomers.

25 Preferably the one or more monomers are selected from the group consisting of monoene monomer. It will be understood by those skilled in the art that by the term "monoene monomer" it is meant a monomer having a single reactive double bond.

The preferred monoene monomer(s) or mixtures thereof include vinyl monomers of structure

30 $\text{CH}_2 = \text{CHX}$ where X is chosen so as to confer the desired reactivity and solubility.

- 14 -

More preferred monomers include styrene. The amount of monomer will preferably be up to 5 times the total moles of initiator added to the polypropylene (co)polymer. The most preferred range being 1 to 4 times the total moles of initiator added to the polypropylene (co)polymer.

5

The monomer may be added with the polypropylene (co)polymer or it can be added prior to the initiator, with the initiator or subsequent to the initiator. However it is preferred to have the monomer mixed and dispersed into the polymer melt before the initiator has substantially decomposed. The monomer is preferably introduced into the polymer melt directly, either
 10 neat (as a powder or a liquid), dispersed or dissolved in a suitable medium (for example, dissolved in 2-butanone) or adsorbed on polymer pellets or powder which are added as a masterbatch.

Preferred initiators for use in combination with monomers include Dibenzoyl peroxide, o,o'-
 15 Bis(methylbenzoyl) peroxide, p,p'-Bis(methylbenzoyl) peroxide, M,M'-Bis(methylbenzoyl) peroxide, o,m'-Bis(methylbenzoyl) peroxide, o,p'-Bis(methylbenzoyl) peroxide, m,p'-Bis(methylbenzoyl) peroxide, Bis(ethylbenzoyl) peroxide (all isomers), Bis(propylbenzoyl) peroxide (all isomers), Bis(butylbenzoyl) peroxide (all isomers), Bis(pentylbenzoyl) peroxide (all isomers), Bis(hexylbenzoyl) peroxide (all isomers), Bis(heptylbenzoyl) peroxide (all isomers),
 20 Bis(octylbenzoyl) peroxide (all isomers), Bis(nonylbenzoyl) peroxide (all isomers), Bis(methoxybenzoyl) peroxide (all isomers), Bis(ethoxybenzoyl) peroxide (all isomers), Bis(propoxybenzoyl) peroxide (all isomers), Bis(butoxybenzoyl) peroxide (all isomers), Bis(pentoxybenzoyl) peroxide (all isomers), Bis(hexyloxybenzoyl) peroxide (all isomers), Bis(heptyloxybenzoyl) peroxide (all isomers), Bis(octyloxybenzoyl) peroxide (all isomers),
 25 Bis(nonyloxybenzoyl) peroxide (all isomers), Bis(chlorobenzoyl) peroxide (all isomers), Bis(fluorobenzoyl) peroxide (all isomers), Bis(bromobenzoyl) peroxide (all isomers), Bis(dimethylbenzoyl) peroxide (all isomers), Bis(trimethylbenzoyl) peroxide (all isomers), Bis(tert-butylbenzoyl)peroxide (all isomers), Bis(di-tert-butylbenzoyl)peroxide (all isomers), Bis(tert-butoxybenzoyl)peroxide (all isomers), Bis(ditrimethylsilylbenzoyl) peroxide (all isomers),
 30 Bis(heptafluoropropylbenzoyl) peroxide (all isomers), Bis(2,4-dimethyl-6- trimethylsilyl benzoyl)

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peroxide and isomers, 2,2'(dioxycarbonyl) bis - Benzoic acid dibutyl ester, tert-butyl perbenzoate, tert-butyl (methyl)perbenzoate (all isomers), tert-butyl (ethyl)perbenzoate (all isomers), tert-butyl (octyl)perbenzoate (all isomers), tert-butyl (nonyl)perbenzoate (all isomers), tert-amyl perbenzoate, tert-amyl (methyl)perbenzoate (all isomers), tert-amyl (ethyl)perbenzoate (all isomers), tert-amyl (octyl)perbenzoate (all isomers), tert-amyl (nonyl)perbenzoate (all isomers), tert-amyl (methoxy)perbenzoate (all isomers), tert-amyl (octyloxy)perbenzoate (all isomers), tert-amyl (nonyloxy)perbenzoate (all isomers), 2-ethylhexyl perbenzoate, 2-ethylhexyl (methyl)perbenzoate (all isomers), 2-ethylhexyl (ethyl)perbenzoate (all isomers), 2-ethylhexyl (octyl)perbenzoate (all isomers), 2-ethylhexyl (nonyl)perbenzoate (all isomers), 2-ethylhexyl (methoxy)perbenzoate (all isomers), 2-ethylhexyl (ethoxy)perbenzoate (all isomers), 2-ethylhexyl (octyloxy)perbenzoate (all isomers), 2-ethylhexyl (nonyloxy)perbenzoate (all isomers), Bis (tertbutylmonoperoxy phthaloyl) diperoxy terephthalate, Bis (tertamylmonoperoxy phthaloyl) diperoxy terephthalate diacetyl phthaloyl diperoxide, dibenzoyl phthaloyl diperoxide, bis(4 methylbenzoyl) phthaloyl diperoxide, diacetyl terephthaloyl di peroxide, dibenzoyl terephthaloyl diperoxide and Poly[dioxycarbonyldioxy(1,1,4,4-tetramethyl-1,4-butanediyl)] peroxide.

Advantageously initiators may be selected to avoid undesirable by-products. In certain applications, it may be desirable to avoid the use of initiators which generate benzene. For example di toluoyl peroxides (bis methyl benzoyl peroxides) may be used in preference to dibenzoyl peroxide.

The processability and other properties of the product may be improved by a chain scission step following the initial polymer modification step. This may be carried out by:

- a) adding one or more additional initiators with or subsequent to the first initiator addition;
- b) the use of high shear mixing;
- c) the use of high temperatures;
- d) the use combination is of one or more of (a) - (c) above.

This additional step in the production of a polymer enables tailoring the properties of the product to meet the requirements of the desired application. For example, by this two stage

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process it is possible to produce materials with similar melt viscosity to the base polymer but a substantially increased melt strength. Use of the single stage process generally provides both an increase in melt strength and an increase in melt viscosity (see examples)

- 5 One or more additional initiators may be added to the polypropylene (co)polymer during the modification process either with or subsequent to the initiator and monomer addition. The additional initiator is typically added to give chain scission of the polypropylene (co)polymer so as to decrease the melt viscosity and improve the processability of the modified polypropylene (co)polymer. The additional initiator should be introduced to the polymer melt
- 10 after the first initiator or have a sufficiently long half-life relative to the first initiator such that its decomposition can be staged to occur after the initial polymer modification process. In some instances a polypropylene (co)polymer modified in accordance with the present invention may have a MFI < 1 g/10 min. With use of the additional initiator an MFI > 1 g/10 min may be achieved. The additional initiator may be selected from the group consisting of
- 15 2,5-dimethyl-2,5-di(t-butylperoxy)hexane (DHBP), dicumyl peroxide (DCP), t-butyl peroxy-2-ethylhexonate (TBEH), and dilauryl peroxide (DLP) or any other peroxide which may result in the overall chain scission of the polypropylene (co)polymer during melt processing. For example in the absence of the monoene monomers, t-butyl peroxybenzoate or other non-preferred initiators for use in the presence of the monomer may be preferably added as the
- 20 additional initiator. While the improvement in processability through chain scission normally results in some decrease in the melt strength/extensional viscosity of the modified polypropylene (co)polymer, the melt strength/extensional viscosity may still be acceptable, and improved over the unmodified polypropylene (co)polymer.
- 25 It is possible to combine the process of the present invention with other processes of polymer modification or with, for example, the addition of fillers, additives or stabilisers, or blending with other polymers. [which do not substantially interfere with the improved properties afforded by the process of the present invention].

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In the process of the present invention the polypropylene (co)polymer is melt mixed in the presence of initiator and monomer. Melt mixing may be carried out by any convenient means capable of mixing the polypropylene (co)polymer at temperatures above the melting point of the polypropylene (co)polymer.

5

Suitable apparatus for melt mixing the polypropylene (co)polymer include continuous and batch mixers. Suitable mixing equipment includes extruders such as single screw and twin screw extruders, static mixers, cavity transfer mixers and combinations of two or more thereof. It is preferred that the melt mixing is conducted in either a co- or counter- rotating
10 twin screw extruder.

The barrel set temperatures are preferably in the range 80-280 °C. Typical melt temperatures are in the range 170-290 °C.

15 In order to optimise the melt strength/extensional viscosity, the preferred melt temperatures are in the range 160 °C to 220 °C. This range provides optimal properties whilst minimising the amount of chain scission which occurs during processing. However, in some cases it may be desirable to use higher temperatures such as in the venting/discharge sections of single screw or twin screw extruders or to induce some chain scission in order to decrease the
20 molecular weight of the modified polypropylene (co)polymer and improve the processability of the modified polypropylene (co)polymer.

Typically, the die temperatures are in the range 180-290 °C.

25 Preferably the extrusion conditions are adjusted so that the polypropylene (co)polymer, initiator/monomer mixture are conveyed as quickly as possible into the melting/mixing zone to maximise the melt phase reaction (eg for twin screw extruders - high throughput rates, higher screw speeds under starve fed conditions). It is more preferred that the additives are added to and mixed with molten polypropylene (co)polymer to further enhance the melt phase

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reaction. Preferably residence times in the range of from 10 seconds to 5 minutes are selected depending upon the temperature profile, throughput rate and initiator levels. More preferred residence times are in the range of from 15 seconds to 120 seconds.

- 5 Vacuum venting can be applied to remove volatile by-products, solvents and/or excess monomer.

While not wishing to be limited by theory, it is believed that the effectiveness of the present invention is determined by three factors:

10

- (a) The rate and specificity of the reaction of the aroyloxy or the derived phenyl radicals or substituted phenyl radicals with polypropylene, and the monomer if present. It is believed that the aroyloxy, phenyl or substituted aroyloxy or phenyl radicals show less specificity for abstraction of tertiary vs. secondary or primary hydrogens than do, for example, alkoxy or
15 alkyl radicals.

- (b) The initiator half-life. Use of an initiator with a short initiator half-life will generate a locally high concentration of radicals thus increasing the likelihood of radical combination events.

20

- (c) The solubility characteristics of the initiator in the polymer melt.

Without wishing to be bound by theory, peroxides that generate aroyloxy or aryl radicals (for example benzoyloxy, p-toluoyloxy) are preferred over those that generate alkoxy radicals (for
25 example, t-butoxy radical, cumyloxy radical). It is believed and supported in the literature that the latter class of peroxides promote chain scission under the melt mixing conditions. While not wishing to be bound by the mechanism, it is believed that this effect is due to the specificity shown by the alkoxy radicals as opposed to the aroyloxy or aryl radicals generated by the peroxides of structure 1. Furthermore we believe that peroxides which generate both

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alkoxy and aryloxy or aryl radicals (for example, t-butyl perbenzoate) show intermediate behaviour. It is believed that they promote less chain scission than peroxides which generate only alkoxy radicals (for example, dialkyl peroxides) when used alone and can be used to advantage in systems where a monomer coagent is employed. Preferred peresters are thus
5 those which generate alkoxy radicals which are not active in hydrogen abstraction (for example t-~~amyl~~-perbenzoate).

Similarly, it is believed, without wishing to be bound by theory, that the effectiveness of the monomer is determined by:

10

(a) The solubility of the monomer in the polymer melt. For example, styrene is known to be soluble in molten polypropylene.

(b) The reactivity of the monomer towards polypropylene derived radicals.

15

(c) The propensity for the radical formed by addition of monomer to give combination or addition (which leads to branch or crosslink formation) vs. disproportionation or hydrogen abstraction. It is known that the benzylic radicals give predominantly combination and have low (with relation to other radicals) tendency to abstract hydrogen.

20

Other initiators and monomers that meet the above criteria may also be used to advantage in the present invention.

Surprisingly, the process of the present invention results in a polypropylene (co)polymer with
25 substantially increased melt strength. We have found that it is possible with the present invention to obtain a polypropylene (co)polymer which has a melt strength at least 25% greater than the melt strength of the base polymer. We have also found that it is possible to obtain an increase in melt strength of greater than 100% for a number of the polypropylene (co)polymers produced in accordance with the process of the present invention. Increases in

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melt strength were assessed using a Gottfert-Rheotens melt strength tester operated with a roller acceleration of 1.2 cm/sec² measuring the melt strength of a 2 mm strand of molten polypropylene (co)polymer (melt temperature of 210°C) which is fed to the Gottfert tester at ~4 g/min.

5

In a further aspect of the present invention there is provided a modified polypropylene (co)polymer produced according to the process described herein, wherein said modified polypropylene (co)polymer preferably has a melt strength at least 25 %, and more preferably at least 100 %, greater than the unmodified polypropylene (co)polymer.

10

The polypropylene (co)polymers produced according to the process of the present invention also may provide a significant increase in long-chain branching. Long-chain branching may be assessed by the Dow Rheology Index. Advantageously, the modified polypropylene (co)polymers may demonstrate a Dow Rheology Index (DRI) of greater than 1, preferably
15 at least 2 and most preferably greater than 50.

The process of the present invention may also be used to increase the melt elasticity of a polypropylene (co)polymer.

20 Advantageously, the process of the present invention also provides a means to alter the molecular weight, molecular weight distribution and/or degree and length of branching of polypropylene, ethylene-propylene copolymers, and analogous α -olefin copolymers with or without altering the melt strength of said polymers by melt processing.

25 The process of the present invention may provide a means to generally increase the molecular weight and broaden the molecular weight distribution and/or introduce branching of the polypropylene (co)polymer. This will not always equate to significant increases in the melt strength or extensional viscosity of the polymer that is being modified eg modification of a lower molecular weight polymer to broaden the molecular weight and/or induce shorter

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branches. Such a product may not necessarily demonstrate a high melt strength, but may demonstrate other desirable properties, for example improved filler uptake, mechanical properties, surface properties, thermal and morphological properties.

- 5 The modified polypropylene (co)polymer produced by the process of the present invention may be used either neat or blended with another polymer or other additives to provide the desired balance of properties in the polymer blend.

The modified polypropylene (co)polymers and blends may be used in a wide variety of
10 applications including thermoforming, blow moulding, tube or pipe extrusion, blown films, foams and extrusion coating.

The present invention may also be used in the recycling of waste polypropylene or materials containing waste polypropylene.

15

The increased melt strength of the modified polypropylene (co)polymers renders these (co)polymers more suitable for use in thermoforming applications. The modified polypropylene (co)polymers may be used to thermoform containers such as margarine tubs. The benefits of this invention include that the polypropylene (co)polymers and blends
20 containing same provide a wider temperature processing window than conventional isotactic polypropylene. The modified polypropylene (co)polymers may also be used in large part thermoforming such as in the production of refrigerator liners and the like where conventional isotactic polypropylene is unsuitable.

- 25 The modified polypropylene (co)polymers produced in accordance with the present invention are suitable for blow moulding and we have found that they can be more readily blow moulded into containers. Furthermore, the increased melt strength makes it possible to produce large blow moulded parts through the use of the high melt strength modified polypropylene (co)polymer. Thus components currently made by rotational moulding may

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now be produced by blow moulding using the modified polypropylene (co)polymer of the present invention.

Profile extrusion for example tube or pipe extrusion, using the modified polypropylene (co)polymer has been found to produce a more consistent product than conventional isotactic polypropylene.

Blown films made of polypropylene are generally blown downwards using relatively expensive equipment. The modified polypropylene (co)polymers of the present invention have sufficient melt strength for them to be able them to be blown upwardly using conventional polyethylene type film blowing equipment which is less expensive and generally more convenient to operate. Advantageously the modified polypropylene (co)polymers of the present invention may be used in the production of blown films.

The modified polypropylene (co)polymers of the present invention may also be foamed with a wider processing window than for conventional polypropylene. Either a physical or chemical blowing agent may be used. It is preferred to use carbon dioxide as a physical blowing agent to produce foams having a fine closed cell structure. Foamed pellets may be subsequently moulded to form components for use in a variety of applications such as automotive door trims, rooflinings, dash boards, bumpers and the like. Applications such as in foamed packaging are also possible, including thermoformed containers, insulating cups and the like.

Waste polypropylene or waste streams containing a significant proportion of polypropylene are presently difficult to recycle as conventionally a high degree of chain scission results from the recycling process. The process of the present invention may be used to upgrade recycled streams containing polypropylene by increasing the overall mechanical properties of the recycled polypropylene by the addition of initiator and monomer in accordance with the present invention.

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The present invention will now be described with reference to the following non-limiting examples. Described hereunder are the measurement techniques used in the examples and a full description of the process conditions employed. Comparative Examples are labelled CE-n.

5

Melt Strength Measurement

Melt strengths were measured on a "Rheotens" Melt Strength Tester, Type 010.1, supplied by Gottfert Werkstoff-Prufmaschinen GmbH of Buchen, Germany. This test involves drawing
10 an extruded strand of polymer vertically into the nip between two counter-rotating nip rollers. The strand was extruded using a Brabender Plasticord single screw extruder of screw diameter 19mm and length to diameter ratio (L/D) of 25. The extrudate exited via a right angle capillary die (2mm diameter). The temperature profile used was uniform along the length of the barrel of the extruder and the die and was set at 190°C. The nip rollers are mounted on
15 a balance arm which allows the force in the drawing strand to be measured. The velocity of the nip rolls is increased at a uniform acceleration rate. As the test proceeds, the force increases until eventually the strand breaks. The force at breakage is termed the "melt strength".

20 While there is no internationally-established standard set of test requirements for melt strength testing, comparative melt strength values obtained under the given set of test conditions provide a quantitative determination of the increase in melt strength used in the patent. The test conditions used were: die temperature 190°C, extruder output rate ~4 g/min, acceleration rate 1.2 cm/sec², draw distance 210 mm, matt finish steel rollers.

25

Dow Rheology Index

The Dow Rheology Index (DRI) is believed in the art to be a measure of the long chain branching in a polymer. It is expressed as the deviation of a viscosity parameter obtained

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from shear rheology measurements on a "branched" polymer compared with that for a linear polymer. The branched polymers have lower values of the viscosity parameter than the linear polymers (for a given relaxation parameter). The parameters are obtained by fitting the Cross model to the shear viscosity flow curves. The DRI method has been described by Lai, Plumley, Butler, Knight and Kao in a paper in SPE ANTEC '94 Conference Proceedings (pp1814-1818) - "Dow Rheology Index (DRI) for Insite Technology Polyolefins (ITP): Unique Structure-Processing Relationships".

Dynamic Rheology Tests

10

The dynamic rheology tests were performed on a Rheometrics Dynamic Stress Rheometer SR200. Test conditions were: parallel plates, temperature 190°C, frequency range 0.01 to 100 rad/sec, and 3-4% strain, in a nitrogen atmosphere to prevent degradation. G' is the storage modulus representing the elasticity of the polymer melt, G'' is the loss modulus which represents the viscous component of the deformation. The polydispersity index is 10 to power 5 divided by the crossover modulus, which is the value of $G' = G''$ when the G' and G'' curves crossover - it is believed to be a measure of MWD. The higher G' , the greater elasticity in the polymer and the higher the MW.

20 MFI

Melt flow indexes (MFI) were measured at 230°C with a 2.16 kg load according to ASTM 1238.

25 Drop Times

The drop times were determined by measuring the time taken for the polypropylene strand (cut at the die face) to drop from the die of the extruder to the floor. The die of the JSW twin screw extruder was 1140 mm above the floor. The drop time test combines the effects of

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melt viscosity, extensional viscosity, chain entanglement (as shown by die swell), and elasticity (as shown by the tendency resist neck formation). Higher melt viscosity polypropylene polymers had drop times which incorporated some additional effect due to prolonged cooling of the slower moving (falling) molten strand.

5

GPC

GPC molecular weights were determined using a Waters 150C high temperature GPC unit. 1,2,4-trichlorobenzene was used as the solvent, eluting through two Styragel HT6E linear
10 columns. The oven temperature was set at 140°C and the pump flow rate was 1.0ml/min.

Calibration was performed using narrow polydispersity polystyrene standards. All molecular weights quoted as polystyrene equivalents.

Mn= number average molecular weight

15 Mw= weight average molecular weight

Mz= viscosity average molecular weight

Mp= peak molecular weight

Twin Screw Extruder

20

The twin screw extruder used in the examples was a JSW TEX-30 with a 30 mm screw diameter and an overall L/D of 42 [comprising ten temperature controlled barrel sections (L/D 3.5, temperatures between 120 and 230°C as specified in Table 1), three unheated sampling/monitoring blocks (L/D 1.167) and a cooled feed block (L/D 3.5)) equipped with
25 two JSW TTF20 gravimetric feeders, one K-Tron KQx gravimetric additives feeder and a volumetric liquid addition pump (Fuji Techno Industries model HYM-03-08)]. The extruder was operated in either co-rotating (intermeshing self wiping) or counter rotating (intermeshing non-self wiping) modes with a throughput rate of between 5 and 20 kg/hr and screw speeds of between 100 and 400 rpm as specified in Table 1. The melt temperature and

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pressures were monitored at three points along the barrel and in the die.

Table 1 Operating conditions

	Conditions	Screw Speed	Feed Rate	Temperature Profile
		(rpm)	(kg/hr)	(°C)
5	A	265	20	150°C, 175°C (by 10)
	B	265	20	180°C, 200°C (by 3), 220°C (by 7)
	C	150	5	120°C, 130°C (by 4), 180°C (by 6)
	D	265	20	140°C, 150°C (by 10)
	E	265	20	180°C, 200°C (by 4), 230°C, 240°C, 250°C, 260°C, 270°C, 280°C.
10	F	400	20	180°C, 220°C (by 10)
	G	265	20	80°C, 120°C, 140°C, 160°C, 170°C, 180°C, 200°C (by 5)
	H	150	5	80°C, 120°C, 140°C, 160°C, 170°C, 180°C, 190°C (by 3), 200°C (by 2)
	I	265	20	80°C, 120°C, 140°C, 160°C, 170°C, 180°C, 190°C (by 3), 200°C (by 2)
	J	250	20	150°C, 170°C (by 3), 180°C, 200°C, 220°C (by 5)

15

- The temperatures in the table refer to sections of the barrel of the extruder that are capable of independent temperature control. The first ten temperatures are barrel section temperatures and the last temperature indicates the temperature of the die.

20

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Table 2 - Die configuration

	Condition	Die Description
	1	Large 3 hole strand die - 6 mm holes
	2	Small 3 hole strand die - 4 mm holes
5	3	Large 2 hole strand die - 6 mm holes
	4	Single hole Brabender die- 10mm hole

Table 3 - Means of modifier addition

	Condition	Die Description
10	α	Modifier added at block 4 in 2-butanone carrier solvent
	β	Modifier added at block 4 in xylene carrier solvent
	γ	Modifier coated onto PP powder - pre tumble blended
	δ	Modifier coated onto PP powder masterbatch

- 15 The overall extruder configuration and modifier conditions may be recited, for example, as condition: A1 δ .

Solvent Addition of Modifiers

- 20 The initiator, and monomer if present, was introduced as a solution in 2-butanone or xylene. The concentration of the initiator varied from 5.6% wt/wt to 8.5% wt/wt. The benzoyl peroxide and the di-toluol peroxides were both powders wetted with 25% (wt/wt) water. The monomer was present in an amount between 4 to 10% wt/wt solvent.
- 25 Increased levels of initiator were generally added by increasing the amount of solution added to the polymer melt. The additional peroxides (if any) were added with the initiator in the carrier solvent.

Solventless Addition of Modifiers

- 30 t-Butyl peroxybenzoate is a liquid. The solventless modification of the polymer was achieved by absorbing the initiator onto powdered polymer or blending it with powdered polymer at

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concentrations ranging from 5% wt/wt to 10% wt/wt to form a masterbatch. The masterbatch was added to the extruder in varying feed rates to alter the amount of additives. The amount of polymer feed was adjusted accordingly to give constant overall feed rate.

- 5 The stabilisers were also added as a masterbatch. The amount of stabiliser was generally kept constant at 0.33%wt/wt Irganox 1010 and 0.17%wt/wt Irgaphos 168 in the total composition.

The main polymer feed was added as either powder or pellets.

10 Single Screw Extruders

Killion

- The Killion single screw extruder used in the examples was a segmented single screw extruder
15 of L/D=40 (11 barrel sections , 10 heated) and screw diameter of 31.75 mm.

Polypropylene powder, stabilisers (0.33%wt/wt Irganox 1010, 0.17%wt/wt Irgaphos 168 in total) and initiator were added to the feed throat of the single screw extruder via a twin screw K-Tron volumetric feeder.

20

Alternatively, the polypropylene powder and stabilisers were added via the K-Tron feeder and polypropylene powder, stabilisers and the modifiers were added as a master batch via a single screw APV Accurate volumetric feeder. The masterbatch contained 7.5%wt/wt benzoyl peroxide (prepared using a dispersion of benzoyl peroxide containing 25%wt/wt water).

25

The output of the extruder was ~1.5 kg/hr using a screw speed of 30rpm. The set barrel temperature was either (i) a flat 220°C with each barrel section and the die set at a temperature of 220°C or (ii) 230°C/190°C with the first six melting sections of the barrel set at 230°C and the next four metering sections of the barrel and the die set at 190°C. The melt

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temperature varied from 220 to 260°C.

Brabender

- 5 The Brabender single screw extruder used was a single screw extruder of L/D=25 (4 Barrel sections), compression ratio 2.5:1 and screw diameter of 19 mm. The die was a 4 mm rod die.

The screw speed of the extruder was 20 rpm. The set barrel temperature was 140°C, 170°C,
10 180°C, 180°C. Residences time: Start 3 min 40 sec; Middle 4 min 35 sec; and End 7 min 30 sec.

Polypropylene powder either as cryoground pellets or ex-reactor powder was mixed with the modifiers and added to the feed throat, either flood feed or by a Brabender single screw
15 volumetric feeder.

The following commercial polypropylene (co)polymers were used in the examples. The properties of the (co)polymers are shown in Table 4 below.

20 Table 4: Comparative data for a grade of high melt strength PP and conventional PP grades.

Example	Polymer	Polymer Description	MFI 2.16kg @ 230°C	Melt Strength cN
Control 1	Montell PF814	High melt strength polypropylene homopolymer	3	18
Control 2	Montell JE6100	Extrusion grade polypropylene homopolymer	3	3

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	Control 3	ICI Australia GYM 45	Injection moulding grade of polypropylene homopolymer	14	1.8
	Control 4	ICI Australia GWM 22	Extrusion grade of polypropylene homopolymer	4	2.8
	Control 5	ICI Australia PXCA 6152	Thermoforming grade of polypropylene homopolymer	0.8	6
	Control 6	ICI Australia LYM 120	Injection moulding grade of propylene/ethylene copolymer	14	1.4
5	Control 7	Montell 6501	Ex-reactor grade of injection moulding polypropylene homopolymer	4.1	~3
	Control 8	Montell KM6100	Extrusion grade of polypropylene homopolymer	~3.5	-
	Control 9	Montell KMT6100	Extrusion grade of polypropylene homopolymer	~3.5	-
	Control 10	Montell KM6100	Ex-reactor grade of polypropylene powder homo- polymer- unstabalized	~3.5	-

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* Melt strength and MFI were measured for a particular batch and we have found actual values vary up to 20% of these values.

Examples 1 to 5

5

GYM45 was modified in accordance with Table 5 below. GYM45 is a low molecular weight/higher MFI injection moulding grade of polypropylene homopolymer.

Table 5:

10	Example	Conditions	BPO	Styrene	Motor	Die	Drop	MFI	Melt Strength
			(wt%)	(wt%)	Current (amps)	Temp. (°C)	Time (secs.)	2.16kg @ 230°C	(cN)
	Control	-	-	0	-	-	-	14	1.8
	3								
	CE 1	B1 α	0	0	13	231	8	12.2	1.5
	1	B1 α	0.36	0	13	229	9.7	14.4	1.9
15	2	B1 α	0.7	0	13	229	13.5	14.4	2.3
	3	B1 α	0.95	0	13	230	17.1	12.5	3
	4	D3 α	1.0	0	19	197	23	9.7	4
	5	D4 α	0.34	0.45	21	179	22.9	9.1	6.9

20

Examples 6 to 18

GWM22 was modified in accordance with Table 6. GWM22 is an intermediate molecular weight/medium MFI extrusion grade of polypropylene homopolymer.

25

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Table 6:

	Example	Conditions	BPO (wt%)	Styrene (wt%)	Motor Current (amps)	Die Temp. (°C)	Drop Time (secs.)	MFI 2.16kg @ 230°C	Melt Strength (cN)
	Control 4							4.5	2.8
5	CE 2	B1 α	0	0	16	239	11.3	5	-
	6	B1 α	0.36	0	16	234	15.2	6.3	3
	7	B1 α	0.75	0	17	238	21.8	5.9	4.7
	8	B1 α	1	0	16	239	25.4	5	6.9
	9	B1 α	1.3	0	20	236	25.3	5.6	7.1
10	10	B1 α	0.12	0.16	16	237	8.0	4.15	-
	11	B1 α	0.23	0.31	17	237	11.2	2.8	5
	12	B1 α	0.46	0.61	21	238	14	1.11	-
	13	B1 α	0.69	0.92	21	241	14.2	0.69	18.6
	14	B1 α	1.22	1.63	21	248	-	-	18.6
15	15	E1 α	0.33	0.44	17	281	20	3.6	8.2
	16	C2 α	0.81	4.2	18	203	60	0.69	18.8
	17	E1 α	0.31	0.40	20	275	23	3.1	7.0
	18	E1 α	0.30	0.53	17	277	24	3.4	9.1

20

The increase in complex viscosity of examples 14, 16, 17 and 18 is shown in Figure 1. G' has been plotted against frequency in Figure 2.

The modified polypropylene's of examples 14, 16, 17 and 18 were tested for additional 25 physical properties and it was found that the modified polypropylene's had:

		14	16	17	18	Control 4	
i)	Elasticity	1200	680	40	45	10	G' @ 0.01 rad/s (pa)
	rad/s (Pa)						
ii)	1/Relaxation Time	~0.0013	0.085	15	18	23	Crossover Frequency(rad/sec)

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iii)	Polydispersity Index	222	39	4.4	4.7	3.7	M_w/M_n
iv)	Dow Rheology Index	192	86	2.0	5.6	0	LongChain Branching

5 Examples 19 to 26

PXCA6152 was modified in accordance with Table 7 below. PXCA6152 is a high molecular weight/low MFI thermoforming grade of polypropylene homopolymer.

10 Table 7:

	Example	Conditions	BPO (wt%)	Styrene (wt%)	Motor Current (amps)	Die Temp. (°C)	Drop Time (secs.)	MFI 2.16kg @ 230°C	Melt Strength (cN)
15	Control 5							0.8	6
	CE 3	F1 α	0	0	17	251	14.6	1.1	-
	19	B1 α	0.34	0	22	244	25.9	1.3	7.4
	20	B1 α	0.68	0	23	250	22.8	1.1	11.1
	21	B1 α	0.8	0	24	246	30.5	0.8	14
	22	B1 α	1.04	0	24	247	25.3	0.65	17.7
	23	F1 α	0.31	0.41	21	256	24.4	0.42	17.5
	24	F1 α	0.47	0.63	21	264	25	0.31	-
20	25	F1 α	0.55	0.73	23	269	-	0.40	-
	26	F1 α	0.71	0.95	22	259	25	0.35	21.3

The modified polypropylene of example 22 was tested for additional physical properties and it was found that the modified polypropylene had:

- i) Elasticity 200 G' @0.01 rad/s (Pa)
- ii) 1/Relaxation Time 7.1 Crossover frequency (rad/sec)

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iii)	Polydispersity Index	3.9	M_w/M_n
iv)	Dow Rheology Index	10	Long Chain Branching

The DRI of the base polypropylene material, PXCA 6152 (an unbranched polypropylene) was expected to be 0. The DRI of the modified polypropylene demonstrates a significant degree of long chain branching.

Examples 27 to 33

10 LYM120 was modified in accordance with Table 8 below. LYM120 is a low molecular weight/higher MFI injection moulding grade of PP copolymer.

Table 8:

Example	Conditions	BPO	Styrene	Motor	Die	Drop	MFI	Melt	
		(wt%)	(wt%)	Current (amps)	Temp. (°C)	Time (secs.)	2.16kg @ 230°C	Strength (cN)	
15	Control 6						12.2	1.4	
	27	D2 α	0.68	0	13	182	19.3	13.1	2.3
	28	A4 α	1.08	0	19.5	200	31	9	4.2
	29	A2 α	0.33	0.44	18	202	28	5.8	7.4
	30	D2 α	0.32	0.42	23	185	46.5	3.8	9.0
20	31	A4 α	0.42	0.55	19.5	204	31.1	6.5	11.2
	32	A4 α	0.62	0.83	20	201	36.8	-	11.9
	33	A4 β	0.34	0.45	16	199	25.1	-	4.3

25 Examples 34 to 42

Ex-reactor GYM45 powder was modified according to Table 9 below. GYM45 is a low molecular weight/higher MFI injection moulding grade of polypropylene homopolymer. The polypropylene was stabilized with Irganox 1010 (0.33 wt%) and Irgaphos (0.17 wt%). The
30 modifiers and stabilizers were added to the twin-screw extruder at the feed throat.

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Table 9:

Example	Conditions	BPO (wt%)	Styrene (wt%)	Motor Current (amps)	Die Temp. (°C)	Drop Time (secs.)	MFI 2.16kg @ 230°C	Melt Strength (cN)
Control	-	-	-	-	-	-	14	1.8
3								
5	CE 4	H3δ	0	7	209	21.5	11.3	1.7
	34	I3δ	0.38	14	209	12.1	13.6	1.9
	36	I3δ	0.75	15	210	15.0	11.8	2.6
	37	I3δ	1.5	15	214	20.6	10.3	5.7
	38	H3δ	0.75	6	209	35.5	17.6	2.1
10	39	H3δ	1.13	8	208	39.5	13.3	2.9
	40	H3δ	1.5	8	208	43.3	9.8	4.4
	41	I3δ	0.15	18	215	16.6	9.6	2.0
	42	I3δ	0.23	16	214	20.5	6.5	5.5
15								

Examples 43 to 49

GYM45 was modified in accordance with Table 10 below. GYM45 is a low molecular weight/higher MFI injection moulding grade of polypropylene homopolymer.

Table 10:

Example	Conditions	Initiator	Initiator (wt%)	Styrene (wt%)	Motor Current (amps)	Die Temp. (°C)	Drop Time (secs)	MFI 2.16kg @ 230°C	
25	CE 5	A3α	-	0	0	18	192	11.8	12.8
	43	A3α	BPO	0.12	0.16	16	197	14	14.4
	44	A3α	BPO	0.21	0.28	17	200	18.8	9.8
	45	A3α	BPO	0.41	0.55	20	206	27.6	5.6
	46	A3α	BPO	0.62	0.83	22	208	32.2	3.6
	CE-6	A3α	DHBP	0.33	0.09	14.5	191	4.6	55.
30	CE-7	A3α	DHBP	0.60	0.17	16	190	4	117

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	CE-8	A3 α	DHBP	0.90	0.28	14.5	190	3.9	132
	CE-9	A3 α	TBEH	0.33	0.34	16	191	9.9	18.3
	CE-10	A3 α	TBEH	0.60	0.62	17	192	10	19.5
	CE-11	A3 α	TBEH	0.90	0.93	17	193	10.6	17.2
5	47	A3 α	TBPB	0.30	0.30	15	194	7.8	58.3
	48	A3 α	TBPB	0.68	0.70	17	198	14	47.3
	49	A3 α	TBPB	0.89	0.91	19	199	15.6	38.5
	CE-12	A3 α	DCP	0.08	0.09	14.5	192	4.3	48.5
	CE-13	A3 α	DCP	0.17	0.17	15	191	3.9	64.7
10	CE-14	A3 α	DCP	0.25	0.25	15	191	3.7	90.3
	CE-15	A3 α	DLP	0.33	0.33	15	190	11.2	16.5
	CE-16	A3 α	DLP	0.63	0.64	15	190	11.1	15.1
	CE-17	A3 α	DLP	0.92	0.93	15	190	11.1	18.0

15

Examples 50 to 54

LYM120 was modified in accordance with Table 11 below. LYM120 is a low molecular weight/higher MFI injection moulding grade of polypropylene copolymer.

20

Table 11:

Example	Conditions	BPO	Motor Current	Die	Drop	MFI	Melt Strength
		(wt%)	(amps)	Temp (°C)	Time (secs.)	2.16kg @ 230°C	(cN)
Control 6						12.2	1.4
CE 18	H3 δ	0	7	209	18.4	11.3	1.2
25	52	H3 δ	0.75	8	210	50.5	6.9
	53	H3 δ	1.13	8	210	47.0	6.6
	54	I3 δ	1.5	14	217	18.5	3.8
							6.1

30

Examples 55 to 61

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LYM120 was modified in accordance with Table 12 below. LYM120 is a low molecular weight/higher MFI injection moulding grade of polypropylene copolymer.

Table 12:

	Example	Conditions	Initiator	Initiator (wt%)	Styrene (wt%)	Motor Current (amps)	Die Temp. (°C)	Drop Time (secs)	MFI 2.16kg @ 230°C	Melt Strength (cN)
5	CE-19	A3 α	BPO	0.00	0.00	16	195	9.9	12.8	1.1
	55	A3 α	BPO	0.12	0.16	15	197	13.9	10.9	
	56	A3 α	BPO	0.21	0.28	17.5	201	17.7	7.50	
	57	A3 α	BPO	0.41	0.55	20	208	25.8	4.4	11.5
10	58	A3 α	BPO	0.62	0.83	21	209	26.5	2.9	
	CE-20	A3 α	DHB	0.08	0.09	13.5	191	4.9	52	
			P							
	CE-21	A3 α	DHB	0.16	0.17	14	190	5.3	79	
			P							
	CE-22	A3 α	DHB	0.28	0.30	14.5	190	5.6	114	
			P							
	CE-23	A3 α	TBEH	0.31	0.32	14	192	8.6	17.8	
15	CE-24	A3 α	TBEH	0.62	0.64	14	192	9	17.4	
	CE-25	A3 α	TBEH	0.98	1.01	14	192	9.6	15.4	
	59	A3 α	TBPB	0.30	0.31	14	196	4.6	33.8	
	60	A3 α	TBPB	0.61	0.62	17	200	14.6	32.9	
	61	A3 α	TBPB	0.93	0.95	17	202	15.6	23.1	
20	CE-26	A3 α	DCP	0.08	0.09	13	192	5	38.6	
	CE-27	A3 α	DCP	0.17	0.17	13.5	190	5.5	57.6	
	CE-28	A3 α	DCP	0.27	0.28	14	190	6.2	65.9	
	CE-29	A3 α	DLP	0.31	0.31	15	191	9.9	15.7	
	CE-30	A3 α	DLP	0.64	0.65	13.5	190	9.8	14.8	
25	CE-31	A3 α	DLP	1.00	1.01	13	190		14.8	

Examples 62 to 73

LYM120 was modified in accordance with Table 13 below. LYM120 is a low molecular weight/higher MFI injection moulding grade of polypropylene copolymer.

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Table 13:

Example	Conditions	Initiator	Initiator (wt%)	Styrene (wt%)	Motor Current (amps)	Die Temp. (°C)	Drop Time (secs)	MFI 2.16kg @ 230°C	Melt Strength (cN)
5	CE-32	A3δ	-	0	14.5	200	10	12.4	1.1
	62	A3δ	BPO	0.11	0	14	199	11.5	1.1
	63	A3δ	BPO	0.23	0	16.5	201	15.1	9.0
	64	A3δ	BPO	0.45	0	16.5	203	20.6	6.6
	65	A3δ	BPO	0.68	0	17.5	206	20.6	5.4
10	66	A3δ	BPO	1.13	0	18	206	18.6	5.7
	CE-33	A3δ	DLP	0.31	0	13.5	198	9.3	13.6
	CE-34	A3δ	DLP	0.59	0	14	199	8.8	14.4
	CE-35	A3δ	DLP	0.89	0	13	196	8.9	15.0
	CE-36	A3δ	TBPB	0.07	0	12	189	5.1	28.7
15	CE-37	A3δ	TBPB	0.15	0	12	190	5.3	31.0
	CE-38	A3δ	TBPB	0.29	0	10.5	185	6.3	92.0
	CE-39	A3δ	TBPB	0.59	0	11	186	11.2	102.0
	67	A3δ	BPO	0.11	0.15	17.5	205	19.1	5.7
	68	A3δ	BPO	0.23	0.30	19.5	210	25	4.3
20	69	A3δ	BPO	0.45	0.6	21.5	209	27.8	2.1
	70	A3δ	BPO	0.90	1.2	23.5	210	26.8	1.3
	CE-40	A3δ	DLP	0.30	0.3	14.5	199	9.4	14.1
	CE-41	A3δ	DLP	0.59	0.6	13	197	9.3	17.2
	CE-42	A3δ	DLP	0.89	0.9	13	197	9.8	16.1
25	71	A3δ	TBPB	0.29	0.3	13.5	196	11.4	20.4
	72	A3δ	TBPB	0.59	0.6	16.5	200	20.2	12.9
	73	A3δ	TBPB	1.18	1.2	17	202	18.8	13.9

Examples 74 to 77

30

GYM45 was modified in accordance with Table 13 below. GYM45 is a low molecular weight/higher MFI injection moulding grade of polypropylene homopolymer.

Table 13

	Example	Conditions	Initiator	Initiator (wt%)	Motor Current (amps)	Die Temp. (°C)	Drop Time (secs)	MFI 2.16kg @ 230°C
5	CE 43	A3 α	-	0	18	192	11.8	12.8
	74	A3 α	BPO	0.23	16.5	196	10.35	17.0
	75	A3 α	BPO	0.45	17	199	11.8	17.3
	76	A3 α	BPO	0.73	17	200	15.9	15.4
	77	A3 α	BPO	0.96	18	202	17.3	14.9
10	CE-44	A3 α	DHBP	0.08	15	191	3.6	96
	CE-45	A3 α	DHBP	0.17	14.5	190	3.1	169
	CE-46	A3 α	DHBP	0.29	13	188	2.3	>100
	CE-47	A3 α	DHBP	0.30	13	188	2.2	>200
	CE-48	A3 α	DHBP	0.50	13.5	186	1.9	>200
15	CE-49	A3 α	DHBP	0.57	12	186	1.9	>100
	CE-50	A3 α	TBEH	0.30	17	197	8.8	12.0
	CE-51	A3 α	TBEH	0.64	14	188	7.2	24.4
	CE-52	A3 α	TBEH	0.98	14	189	6.8	25.6
	CE-53	A3 α	TBPB	0.31	13	186	2.6	95
20	CE-54	A3 α	TBPB	0.64	13	184	2.1	238
	CE-55	A3 α	TBPB	1.03	12	184	1.9	>250
	CE-56	A3 α	DCP	0.08	16	192	3.8	47.1
	CE-57	A3 α	DCP	0.17	14	189	2.9	121.3
	CE-58	A3 α	DLP	0.32	16	191	10.9	14.5
25	CE-59	A3 α	DLP	0.64	16	190	10.6	17.9
	CE-60	A3 α	DLP	0.95	16	189	10.4	16.6

Examples 78 to 82

30 LYM120 was modified in accordance with Table 14 below. LYM120 is a low molecular weight/higher MFI injection moulding grade of polypropylene copolymer.

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Table 14:

Example	Conditions	Initiator	Initiator (wt%)	Motor Current (amps)	Die Temp. (°C)	Drop Time (secs)	MFI 2.16kg @ 230°C
5	Control 24	A3α	BPO	0	16	195	12.8
	78	A3α	BPO	0.25	15	196	13.4
	79	A3α	BPO	0.47	14	198	13.1
	80	A3α	BPO	0.64	16	198	12.9
	81	A3α	BPO	0.70	17	201	11.9
10	82	A3α	BPO	0.94	18	198	10.6
	CE-61	A3α	DHBP	0.09	14	190	80
	CE-62	A3α	DHBP	0.16	13	188	160
	CE -63	A3α	DHBP	0.26	13	187	250
	CE-64	A3α	TBEH	0.29	15	193	7
15	CE-65	A3α	TBEH	0.60	13	192	19.4
	CE-66	A3α	TBEH	1.02	12	191	22.6
	CE-67	A3α	TBPB	0.30	13	186	85
	CE-68	A3α	TBPB	0.61	12	184	173
	CE-69	A3α	TBPB	0.92	12	184	>250
20	CE-70	A3α	DCP	0.08	16	192	14.6
	CE-71	A3α	DCP	0.17	13	190	107
	CE-72	A3α	DCP	0.25	13	188	131
	CE-73	A3α	DLP	0.32	14.5	191	14.3
	CE-74	A3α	DLP	0.68	15	191	15.8
25	CE-75	A3α	DLP	0.98	14.5	193	21.0

Examples 83 to 92

LYM120 was modified in accordance with Table 15 below. LYM120 is a low molecular weight/higher MFI injection moulding grade of polypropylene copolymer.

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Table 15:

Example	Conditions	Initiator	Initiator (wt%)	Styrene (wt%)	Mole Ratio Sty/Init	Motor Current (amps)	Die Temp. (°C)	Drop Time (secs)	MFI 2.16kg @ 230°C	Melt Strength (cN)	
5	CE-76	A3α	0	0	0	-	16	195	9.9	12.8	1.1
	83	A3α	BPO	0.43	0.19	1.04	16	207	18.5	6.9	-
	84	A3α	BPO	0.41	0.37	2.07	19	207	22.5	4.9	-
	85	A3α	BPO	0.41	0.55	3.11	20	208	25.8	4.4	11.6
	86	A3α	BPO	0.43	0.76	4.14	21	210	25.4	3.9	-
10	87	A3α	BPO	0.45	0.99	5.18	20	205	28.3	4.4	-
	88	A3α	TBPB	0.56	0.19	0.78	13	192	8.9	71	-
	89	A3α	TBPB	0.55	0.37	1.57	14	196	11.0	38	-
	90	A3α	TBPB	0.61	0.62	1.91	17	200	14.6	33	-
	91	A3α	TBPB	0.54	0.73	3.14	16	201	16.0	19.2	-
15	92	A3α	TBPB	0.58	0.97	3.92	19	204	16.2	15.7	-
	CE-77	A3α	DHB	0.27	0.1	0.84	13	190	4.4	187	-
			P								
	CE-78	A3α	DHB	0.25	0.18	1.69	14	191	5.3	125	-
			P								
20	CE-79	A3α	DHB	0.28	0.30	3.04	14.5	190	5.6	114	-
			P								
	CE-80	A3α	DHB	0.27	0.40	3.38	15	193	6.2	116	-
			P								
	CE-81	A3α	DHB	0.28	0.50	4.22	14	192	6.1	118	-
			P								

20

Examples 93 to 97

GYM45 was modified in accordance with Table 16 below. GYM45 is a low molecular weight/higher MFI injection moulding grade of polypropylene homopolymer.

25

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Table 16:

Example	Conditions	Initiator	Initiator (wt%)	Styrene (wt%)	Mole Ratio Sty/Init	Motor Current (amps)	Die Temp (°C)	Drop Time (secs)	MFI 2.16kg @ 230°C
5	CE-82	A3 α	-	0.00	0.00	18	192	11.8	12.75
	93	A3 α	BPO	0.36	1.04	18	202	18.1	11.17
	94	A3 α	BPO	0.41	2.07	18	209	23.5	6.38
	95	A3 α	BPO	0.41	3.11	20	206	27.6	5.62
	96	A3 α	BPO	0.43	4.14	22	209	25.7	4.05
10	97	A3 α	BPO	0.40	5.18	21	207	31.2	4.27

Example 98 to 105

LYM120 was modified in accordance with Table 17 below. LYM120 is a low molecular weight/higher MFI injection moulding grade of polypropylene copolymer.

Table 17:

Example	Conditions	Initiator #1 wt%	Initiator #2 wt%	Mole ratio	Monomer wt%	Monomer ratio	Motor Current (amps)	Drop time secs	Die Te mp °C	MFI	Melt strength (cN)
Initiator #1=BPO, Initiator #2=DHBP, Monomer=Styrene											
98	A3δ	0.43	0.06	9.08	0.57	2.81	19	17	204	6.9	3.8
99	A3δ	0.43	0.11	4.54	0.58	2.57	19	14.8	203	10.5	2.9
100	A3δ	0.43	0.17	3.03	0.58	2.37	18	14	201	16.9	-
Initiator #1=BPO, Initiator #2=TBPB, Monomer=Styrene											
101	A3δ	0.43	0.11	3.07	0.58	2.36	20	18.7	208	5	5.5
102	A3δ	0.43	0.22	1.53	0.58	1.92	17	14.2	204	9.3	4.2
103	A3δ	0.43	0.34	1.02	0.59	1.62	18	14	204	11.6	-
104	A3δ	0.43	0.45	0.77	0.60	1.40	18	11.8	201	19.5	-
105	A3δ	0.43	0.34	1.02	0.94	2.59	21	17	207	7.5	6.3

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Example 108 and 109

Montell 6501 was modified in accordance with Table 19 below on the Killion screw extruder described above.

5

Sample	Barrel Temp (°C)	Extruder Output (kg/hr)	BPO wt%	Styrene wt%	Motor Current (amps)	Die Temp (°C)	Drop Time (secs)	MFI (g/10 min)
Control	-	-	-	-	-	-	-	4.1
7								
CE 84	220	1.4	0	0	6	256	17	4.1
	flat							
10 108	220	1.4	2.1	0.25	6	260	35	2.2
	flat							
109	220	1.4	4.2	0.5	7.5	263	33	0.40
	flat							

Examples 40, 41, 7, 12, 28, 29, 31 and 14

15

GPC molecular weights were determined using a Waters 150C high temperature GPC unit. 1,2,4-trichlorobenzene was used as the solvent, eluting through two Ultrastaygel linear columns. The oven temperature was set at 140°C and the pump flow rate was 1.0 ml/min.

20 Calibration was performed using narrow polydispersity polystyrene standards. All molecular weights quoted as linear polystyrene equivalents.

Mn=number average molecular weight

Mw=weight average molecular weight

25 Mz=viscosity average molecular weight

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Mp=peak molecular weight

Error are quoted as two times the standard deviation between duplicate injections.

5 Table 20:

Exam ple No.	Cond.	BPO (wt%)	Sty (wt%)	MFI (g/10 min)	Melt str (cN)	Mn (g/mo l) x 10 ⁻³	Mw (g/mo l) x 10 ⁻³	Mz (g/mo l) x 10 ⁻³	Mp (g/mo l) x 10 ⁻³
Intermediate Molecular Weight PP Homopolymer (GWM 22)									
10	Contr ol 4	-	-	4.5	2.8	55	295	1200	105
	8	B1 α	1.0	5.0	6.9	80	425	1415	200
	10	B1 α	0.12	0.16	4.15	-	90	405	235
15	11	B1 α	0.23	0.31	2.80	5.0	75	415	195
	12	B1 α	0.46	0.61	1.11	-	70	555	2200
	13	B1 α	0.69	0.92	0.69	18.6	85	575	2200
	110	C2 α	1.50	3.8	-	75	430	1560	160
	111	C2 α	2.23	3.2	-	75	430	1700	150
20	112	C1 α	0.37	0.49	2.22	-	85	565	2035
	113	C1 α	0.60	0.80	1.00	19.4	85	690	2575
	114	C1 α	0.32	1.65	4.50	-	80	505	1835
	115	C1 α	0.47	2.45	1.58	-	90	605	2160
	116	C1 α	0.81	4.19	0.69	18.8	85	675	2610
25	Low Molecular Weight PP Copolymer (PXCA 6152)								
	Contr ol	-	-	12.4	1.4	45	230	720	130

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5									
22	B1 α	1.04		0.65	17.7	110	485	1615	195
27	A2 α	0.33	0.44	-	7.4	65	325	1045	140
61	A2 α	0.41	0.55	4.4	11.5	60	325	1315	125
5 62	A2 α	0.62	0.83	2.9	-	70	460	2435	135
-	D2 α	0.28	0.38	-	-	80	555	2875	160
28	D2 α	0.32	0.44	-	9.0	120	640	4130	140
78	A3 α	0.25		13.4	-	70	315	1330	135
79	A3 α	0.47		13.1	-	65	320	1380	130
10 80	A3 α	0.64		12.9	-	65	360	1975	130
27	D3 α	0.68		13.1	2.3	70	445	1865	140

*Errors in the molecular weight are generally less than 30% of the quoted value, as is usual in high temperature GPC under the conditions employed.

15

Examples 110 to 114

GWM22 and KM6100 were modified in accordance with Table 20 below.

Table 20: Effect of feed throat addition of BPO on the modification of prestabilised PP
20 homopolymer [KM6100 or GWM22] *a*

Example	Conditions	Powder wt%	BPO (wt%)	Motor Current (amps)	Drop Time (sec)	Die Temp °C	MFI (g/10 min.)	Melt Str. (cN)
Prestabilised PP homopolymer GWM 22								
Control							~4	2.8
25 4								
110	J3 δ	8.8	0.92	25	18	244	3.9	11.0
111	J3 δ	13.3	1.40	30	19	248	2.6	8.5
Prestabilised PP Homopolymer KM6100								

- 47 -

Control							~3.5	2.5	
8									
5	CE-85	J3δ	0	0	27	11	240	3.7	2.5
	112	J3δ	2.0	0.41	26	15	235	4.9	3.9
	113	J3δ	3.9	0.81	27	19	241	3.3	6.7
	114	J3δ	6.2	1.28	28	19	241	2.7	9.5

a: BPO added to pellet feed in PP powder derived from cryoground prestabilised PP pellets

10

Examples 115 to 117

KMT6100 was modified in accordance with Table 22 below. KMT6100 is a prestabilised PP
15 copolymer.

Table 22: Effect of feed throat addition of BPO on the modification of prestabilised PP
copolymer [KMT6100]

Example	Conditions	Powder	BPO	Motor	Drop	Die	MFI	Melt Str.	
		wt%	(wt%)	Current	Time	Temp	(g/10 min.)	(cN)	
		<i>a</i>		(amps)	(sec)	°C			
20	Control						~3.5	2.0	
	9								
25	CE-86	J3δ	0	0	24	8	231	4.4	2.0
	115	J3δ	1.9	0.40	25	10	233	5.4	1.9
	116	J3δ	3.0	0.81	24	13	235	4.4	3.1
	117	J3δ	5.9	1.22	29	15	237	3.0	4.5

a: BPO added to pellet feed in PP powder derived from cryoground prestabilised PP pellets

30

Examples 118 to 121

KM6100u was modified with para-toluoyl peroxide (PTP) and BPO in accordance with Table 23 below. The KM6100u was stabilized with Irganox 1010 (0.33wt%) and Irgaphos 168 (0.17 wt%) which were added to the main feed throat of the extruder.

Table 23:

Example	Conditions	Peroxide	Peroxide (wt%)	Motor Current (amps)	Drop Time (sec)	Die Temp °C	MFI (g/10 min.)	Melt Str. (cN)	
Control							~3.5	~2.5	
10	10								
	CE-87	J3δ	-	0	22	9	240	5.2	2.7
	118	J3δ	BPO	1.0	22	17	252	5.2	7.2
	119	J3δ	PTP	1.0	21	16	240	5.2	6.8
	120	J3δ	PTP	1.5	22	18	239	3.9	14.2
15	121	J3δ	PTP	2.0		24	243	3.4	14.0

Examples 122 to 128

PXCA6152 was modified with mixed initiator systems in accordance with Table 24 below.

5 Table 24:

Example	Conditions	Init #1 wt%	Init #2 wt%	Mole ratio	Motor Current Init#1/In it #2	Drop Time (sec)	Die Temp °C	MFI (g/10 min.)	Melt Str. (cN)
Control								0.8	6
5									
10	CE-88	0	0	-	22	14	255	0.9	5.1
122	B3 α	0.87	0	-	24	20	257	1.28	14.2
Initiator #1 = BPO, Initiator #2 = DHBP									
15									
123	B3 α	0.87	0.045	23.2	22	19	251	4.0	8.0
124	B3 α	0.87	0.064	16.3	20	16	250	5.4	-

125 B3 α 0.87 0.084 12.4 20 15 248 6.4 0.2

Initiator #1 = BPO, Initiator #2 = TBPB

126 B3 α 0.87 0.006 129.4 22 16 254 3.4 -
 127 B3 α 0.87 0.012 64.7 20 16 249 3.9 0.5
 128 B3 α 0.87 0.019 40.9 20 16 249 6.4 0.1

Examples 129 to 132

15 Cryoground PXCA6152 in the form of a powder was modified with mixed initiator systems according to Table 25 below.

Table 25: Effect of Mixed Initiators on the Modification of PXCA6152 Powder *a* (cryoground pellets)

20

Example	Conditions	Init #1 wt%	Init #2 wt%	Mole ratio Init#1/Init #2	Motor Current (amps)	Drop Time (sec)	Die Temp °C	MFI (g/10 min.)	Melt Str. (cN)
Control								0.8	6
5	CE-89 B3δ	0	0	-	25	13	256	1.0	-
5	Initiator #1 = BPO, Initiator # 2 = DHBP								
129	B3δ	0.86	0	-	23	17	253	1.8	10.8
10	130 B3δ	0.87	0.004		23	17	254	2.1	8.8
	Initiator #1 = BPO, Initiator # 2 = TBPB								
131	B3δ	0.87	0.016		24	18	253	2.6	8.2
15	132 B3δ	0.87	0.026		23	17	253	3.6	7.7

Examples 79 and 85

20 LYM120 was modified in accordance with Table 26 below.

Table 26:

Example	Conditions	BPO wt%	Monomer/ Coagent	Monom er wt%	Motor Current (amps)	Drop Time (sec)	Die Temp °C	MFI (g/10 min.)	Melt Str. (cN)
Control								12.2	1.4
6									
5	CE-76	0	none	0	16	10	195	12.8	1.1
79	A α	0.47	none	0	14	13	198	13.2	-
85	A α	0.41	Styrene	0.54	20	25.8	208	4.4	11.6

10 Examples 133

GYM 22 was modified in accordance with Table 27 below.

Table 27:

Example	Conditions <i>a</i>	BPO wt%	Monomer/ Coagent	Monomer wt%	Motor Current (amps)	Drop Time (sec)	Die Temp °C	MFI (g/10 min.)	Melt Str. (cN)
Control								4.5	2.8
5									
4									
CE-2	B1 α	0	none	-	16	11	239	5	-
6	B1 α	0.36	none	-	16	15	234	6.3	3
133	B1 α	0.34	Styrene	0.45	21	29	250	1.72	-

10

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Examples 134 to 137

Cryoground KM6100 in the form of a powder was modified on a Brabender single screw extruder in accordance with the general description of the Brabender SSE above and Table 28 below. The initiator was added at the feed throat of the SSE along with the stabilizers (0.33 wt% Irganox 1010 and 0.17 wt% Irgaphos 168).

Table 28:

10	Example	Peroxide	Peroxide	MFI
		Type	(wt%)	(g/10 min.)
	Control 8	-	0	3.5
	134	BPO	1	2.9
	135	PTP	0.5	3.8
	136	PTP	1	3.3
15	137	PTP	2	2.0

* PTP- Paratoluol Peroxide (bis paramethyl benzoyl peroxide)

20 Example 138

PXCA6152 was modified in accordance with Table 29 below.

Table 29: Modification of PXCA6152 Pellets

25	Example	Conditions	BPO	Styrene	Motor	Drop	Die	MFI	Melt
			wt%	wt%	Current (amps)	Time (sec)	Temp °C	(g/10 min.)	Str. (cN)
	Control							0.8	6.0
5	CE-94	B3 α	0	-	22	14	255	0.9	5.1
	138	B3 α	0.51	0.68	24	19	279	0.6	21.0

30

The modified PXCA6152 produced according to Example 138 was melt mixed with GYM45 in accordance with Table 30.

Table 30: Blends of the Modified PP with other PP Homopolymers

Example	Conditions	PP#1 (wt%)	PP#1 (wt%)	Motor Current (amps)	Drop time (seconds)	Die Temp °C	MFI	Melt Strength
5	CE-95	A3	Control 5 (5)	18	11	206	9.9	
	CE-96	A3	Control 5 (10)	19	12	202	7.8	
	CE-97	A3	Control 5 (15)	19	13	202	7.0	
	CE-98	A3	Control 5 (20)	19	14	202	5.5	
	CE-99	A3	Control 5 (25)	19	15	202	4.7	2.6
10	139	A3	138 (5)	19	15	199	9.3	
	140	A3	138 (10)	19	16	201	7.4	
	141	A3	138 (15)	19	18	203	6.6	
	142	A3	138 (20)	20	19	204	5.0	
	143	A3	138 (25)	19	20	207	4.7	5.2
15	CE-100	A3	-	20	15	208	5.3	
	144	A3	138 (5)	21	18	207	3.7	
	145	A3	138 (10)	21	19	206	3.5	
	146	A3	138 (15)	22	21	210	2.3	
	147	A3	138 (20)	22	21	212	2.6	
20	148	A3	138 (25)	23	23	213	2.4	7.7

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Examples of Carbon Dioxide Foaming of Modified PP

The equipment used for foaming the polypropylene (from earlier examples) was a tandem extrusion line made up of an Leitritz twin screw extruder (34 mm screw diameter, co-rotating, with 11 barrel sections) connected via a melt pipe to a single screw extruder (43 mm screw diameter). CO₂ was introduced into barrel six of the twin screw extruder. The gassed polymer was then cooled slowly in the single screw extruder.

Example	MFI (g/10 min)	Melt Strength (cN)	Foaming Temp (°C)	Av Foam Density (g/cc)	Av Cell Size (µm)
10 Control 1	3	18	166 to 159	0.058	550
25	0.4	-	169 to 159	0.044	300
31	6.5	11.2	167 to 161	0.051	280

Non high melt strength grades of polypropylene have foam temperature processing windows of less than 1°C.

Foamed examples 13 and 17 both has a fine closed cell structure.

Examples of thermoforming

20

The modified polypropylene produced in Example 69 was extruded on a Welex single screw extruder through a sheet die to produce a sheet 78cm wide and ~1.25 mm thick. The sheet was fed to a Gabler F702 continuous thermoformer to produce margarine tubs. Tubs produced from the modified PP sample had a crush strength of 25 kg after 1 hour. No appreciable sag was noticed of the PP sheet during the process.

Blow Moulding

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The modified polypropylene of Example 5 was blow moulded on Bekum blow moulder fitted with a general purpose polyolefin screw using a 750 ml screw top bottle mould, (radially non symmetrical bottle with waist). The mould temperature was 0°C.

5

The blow mouldability of the modified injection moulding grade of PP was compared against that of a commercial low melt flow index PP homopolymer (ICI GWM110 of MFI = 1.5).

It was found that the modified PP homopolymer (MFI = 9.1 and Melt Strength = 6.9 cN) could be easily blow moulded into 750 ml bottles. Conventional PP of similar MFI could not be successfully blow moulded. The modified PP gave similar performance to an extrusion grade PP of low MFI.

The results are very promising where a higher MFI PP could be used to blow bottles. This possibly opens up the opportunity to produce large blow moulded parts through use of a high melt strength modified PP which has been tailored to have an MFI acceptable to blow moulding (ie 1-2 MFI)

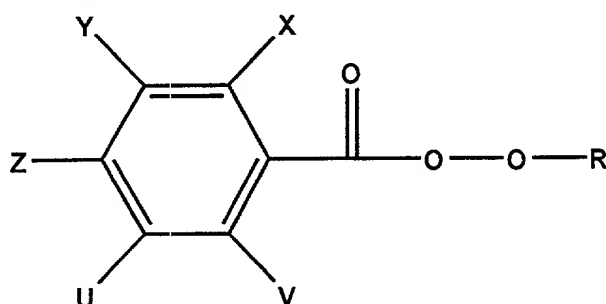
Those skilled in the art will appreciate that the invention described herein is susceptible to variations and modifications other than those specifically described. It is to be understood that the invention includes all such variations and modifications which fall within its spirit and scope. The invention also includes all of the steps, features, compositions and compounds referred to or indicated in this specification, individually or collectively, and any and all combinations of any two or more of said steps or features.

25

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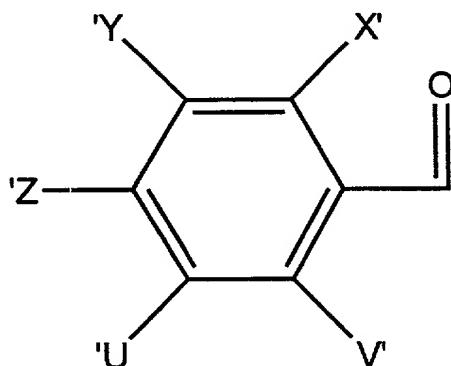
CLAIMS

1. A process for increasing the melt strength and/or the extensional melt viscosity of a polypropylene (co)polymer wherein said process comprises melt mixing the polypropylene (co)polymer in the presence of an initiator wherein said initiator is selected from the group defined by formula 1.



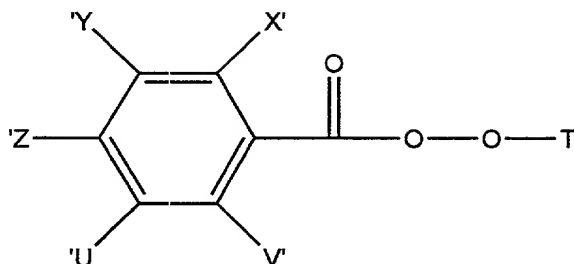
Formula 1

- 10 wherein R is selected from the group consisting of optionally substituted C_1 to C_{18} acyl, optionally substituted C_1 to C_{18} alkyl, aroyl defined by formula 2,



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and groups of formula 3,

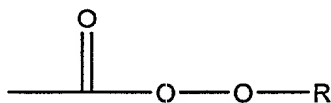


Formula 3

5

wherein U, V, X, Y, Z, U', V', X', Y' and Z' are independently selected from the group consisting hydrogen, halogen, C1-C18 alkyl, C1-C18 alkoxy, aryloxy, acyl, acyloxy, aryl, carboxy, alkoxycarbonyl, aryloxycarbonyl, trialkyl silyl, hydroxy, or a moiety of formula 4,

10



Formula 4

and wherein T is alkylene.

2. A process according to claim 1 wherein the initiator is selected from compounds of formula 6.

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(methoxy)perbenzoate (all isomers), tert-amyl (octyloxy)perbenzoate (all isomers), tert-amyl (nonyloxy)perbenzoate (all isomers), Bis (tertamylmonoperoxy phthaloyl) diperoxy terephthalate, diacetyl phthaloyl diperoxide, dibenzoyl phthaloyl diperoxide, bis(4-methylbenzoyl) phthaloyl diperoxide, diacetyl terephthaloyl di peroxide and dibenzoyl
5 terephthaloyl diperoxide.

11. A process according to claim 10 wherein the initiator is more preferably the initiators are selected from the group consisting of dibenzoyl peroxide, o,o'-Bis(methylbenzoyl) peroxide, p,p'-Bis(methylbenzoyl) peroxide, M,M'-Bis(methylbenzoyl) peroxide, o,m'-Bis(methylbenzoyl)
10 peroxide, o,p'-Bis(methylbenzoyl) peroxide, m,p'-Bis(methylbenzoyl) peroxide.

12. A process according to claim 1 wherein the initiator is used in combination with a monomer.

15 13. A process according to claim 12 wherein the amount of monomer is up to 5 times the total moles of initiator.

14. A process according to claim 12 or claim 13 wherein the monomer is a monoene monomer.

20

15. A process according to claim 12 or claim 13 wherein the monomer is styrene.

16. A process according to claim 12 wherein the initiator is selected from the group consisting of Dibenzoyl peroxide, o,o'-Bis(methylbenzoyl) peroxide, p,p'-Bis(methylbenzoyl)
25 peroxide, M,M'-Bis(methylbenzoyl) peroxide, o,m'-Bis(methylbenzoyl) peroxide, o,p'-Bis(methylbenzoyl) peroxide, m,p'-Bis(methylbenzoyl) peroxide, Bis(ethylbenzoyl) peroxide (all isomers), Bis(propylbenzoyl) peroxide (all isomers), Bis(butylbenzoyl) peroxide (all isomers), Bis(pentylbenzoyl) peroxide (all isomers), Bis(hexylbenzoyl) peroxide (all isomers), Bis(heptylbenzoyl) peroxide (all isomers), Bis(octylbenzoyl) peroxide (all isomers),
30 Bis(nonylbenzoyl) peroxide (all isomers), Bis(methoxybenzoyl) peroxide (all isomers),

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Bis(ethoxybenzoyl) peroxide (all isomers), Bis(propoxybenzoyl) peroxide (all isomers),
 Bis(butoxybenzoyl) peroxide (all isomers), Bis(pentoxybenzoyl) peroxide (all isomers),
 Bis(hexyloxybenzoyl) peroxide (all isomers), Bis(heptyloxybenzoyl) peroxide (all isomers),
 Bis(octyloxybenzoyl) peroxide (all isomers), Bis(nonyloxybenzoyl) peroxide (all isomers),
 5 Bis(chlorobenzoyl) peroxide (all isomers), Bis(fluorobenzoyl) peroxide (all isomers),
 Bis(bromobenzoyl) peroxide (all isomers), Bis(dimethylbenzoyl) peroxide (all isomers),
 Bis(trimethylbenzoyl) peroxide (all isomers), Bis(tert-butylbenzoyl)peroxide (all isomers),
 Bis(di-tert-butylbenzoyl)peroxide (all isomers), Bis(tert-butoxybenzoyl)peroxide (all isomers),
 Bis(ditrimethylsilylbenzoyl) peroxide (all isomers), Bis(heptafluoropropylbenzoyl) peroxide (all
 10 isomers), Bis(2,4-dimethyl-6- trimethylsilyl benzoyl) peroxide and isomers,
 2,2'(dioxycarbonyl) bis - Benzoic acid dibutyl ester, tert-butyl perbenzoate, tert-butyl
 (methyl)perbenzoate (all isomers), tert-butyl (ethyl)perbenzoate (all isomers), tert-butyl
 (octyl)perbenzoate (all isomers), tert-butyl (nonyl)perbenzoate (all isomers), tert-amyl
 perbenzoate, tert-amyl (methyl)perbenzoate (all isomers), tert-amyl (ethyl)perbenzoate (all
 15 isomers), tert-amyl (octyl)perbenzoate (all isomers), tert-amyl (nonyl)perbenzoate (all isomers),
 tert-amyl (methoxy)perbenzoate (all isomers), tert-amyl (octyloxy)perbenzoate (all isomers),
 tert-amyl (nonyloxy)perbenzoate (all isomers), 2-ethylhexyl perbenzoate, 2-ethylhexyl
 (methyl)perbenzoate (all isomers), , 2-ethylhexyl (ethyl)perbenzoate (all isomers), 2-ethylhexyl
 (octyl)perbenzoate (all isomers), , 2-ethylhexyl (nonyl)perbenzoate (all isomers), 2-ethylhexyl
 20 (methoxy)perbenzoate (all isomers), 2-ethylhexyl (ethoxy)perbenzoate (all isomers), 2-
 ethylhexyl (octyloxy)perbenzoate (all isomers), 2-ethylhexyl (nonyloxy)perbenzoate (all
 isomers), Bis (tertbutylmonoperoxy phthaloyl) diperoxy terephthalate, Bis (tertamylmonoperoxy
 phthaloyl) diperoxy terephthalate diacetyl phthaloyl diperoxide, dibenzoyl phthaloyl diperoxide,
 bis(4 methylbenzoyl) phthaloyl diperoxide, diacetyl terephthaloyl di peroxide, dibenzoyl
 25 terephthaloyl diperoxide and Poly[dioxycarbonyldioxy(1,1,4,4-tetramethyl-1,4-butanediyl)]
 peroxide.

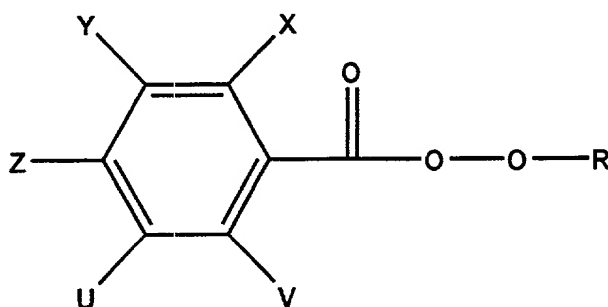
17. A modified polypropylene produced according to any one of the processes of claims
 1, 10 and 12.

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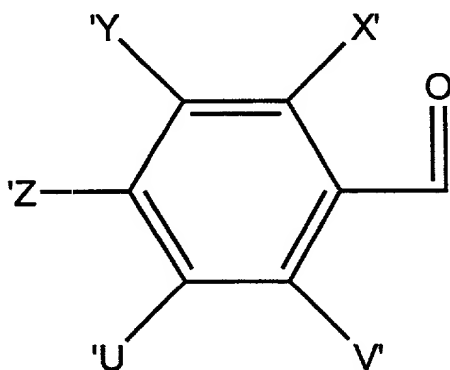
- 65 -

18. A process wherein the modified polypropylene of claim 17 is melt mixed with an unmodified polypropylene to produce a modified polypropylene.

19. A process for modifying an α -olefin polymer wherein said process comprises melt
5 mixing the α -olefin polymer in the presence of an initiator and optionally a monoene monomer wherein said initiator is selected from the group defined by formula 1.

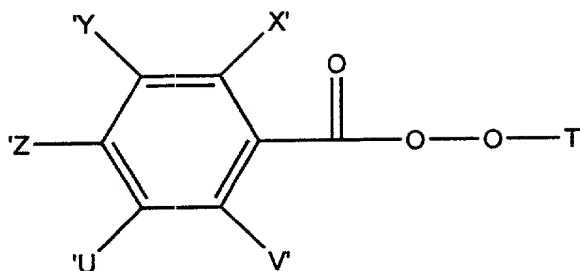
**Formula 1**

10 wherein R is selected from the group consisting of optionally substituted C₁ to C₁₈ acyl, optionally substituted C₁ to C₁₈ alkyl, aroyl defined by formula 2,

**Formula 2**

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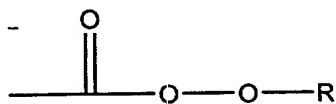
and groups of formula 3,

**Formula 3**

5

wherein U, V, X, Y, Z, U', V', X', Y' and Z' are independently selected from the group consisting hydrogen, halogen, C1-C18 alkyl, C1-C18 alkoxy, aryloxy, acyl, acyloxy, aryl, carboxy, alkoxycarbonyl, aryloxycarbonyl, trialkyl silyl, hydroxy, or a moiety of formula 4,

10

**Formula 4**

and wherein T is alkylene;

and wherein the amount of monomer is 0 to 5 times the total moles of initiator.

15

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PTO/SB/01 (12-97)
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**DECLARATION FOR UTILITY OR
DESIGN
PATENT APPLICATION
(37 CFR 1.63)**

☐ Declaration Submitted with Initial Filing **OR** ☒ Declaration Submitted after Initial Filing (surcharge (37 CFR 1.16 (e)) required)

Attorney Docket Number	US 17017
First Named Inventor	Gary Peeters et al.
COMPLETE IF KNOWN	
Application Number	09 / 600,363
Filing Date	July 14, 2000
Group Art Unit	
Examiner Name	

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

PROCESS FOR INCREASING THE MELT STRENGTH OF
POLYPROPYLENE

the specification of which *(Title of the Invention)*

☐ is attached hereto
OR

☒ was filed on (MM/DD/YYYY) 01/19/1999 as United States Application Number or PCT International
13 August 1999

Application Number PCT/AU99/00036 and was amended on (MM/DD/YYYY) 26 Nov 1999 (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. 119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached?	
				YES	NO
PP1393/98	Australia	01/19/1998	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
PP1392/98	Australia	01/19/1998	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>

☐ Additional foreign application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto:

I hereby claim the benefit under 35 U.S.C. 119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)	<input type="checkbox"/> Additional provisional application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto.

[Page 1 of 3]

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DECLARATION — Utility or Design Patent Application

I hereby claim the benefit under 35 U.S.C. 120 of any United States application(s), or 365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application or PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
PCT/AU99/00036	01/19/1999	

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As a named inventor, I hereby appoint the following registered practitioner(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

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Margaret S. Millikin	38,969	Joanne L. Horn	30,392

☐ Additional registered practitioner(s) named on supplemental Registered Practitioner Information sheet PTO/SB/02C attached hereto

Direct all correspondence to: ☐ Customer Number or Bar Code Label OR ☒ Correspondence address below

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City	Elkton	State	MD	ZIP	21921
Country	US	Telephone	410-996-1646	Fax	410-996-1560

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor:

☐ A petition has been filed for this unsigned inventor

Given Name (first and middle [if any])			Family Name or Surname		
Gary			Peeters		
Inventor's Signature	Gary Peeters			Date	27/9/2000
Residence: City	Victoria	State		Country	AU
Post Office Address	89 Duff Street				
Post Office Address	Cranbourne				
City	Victoria	State		ZIP	3977
				Country	AU

☒ Additional inventors are being named on the 1 supplemental Additional Inventor(s) sheet(s) PTO/SB/02A attached hereto

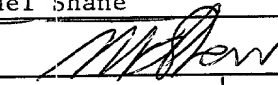
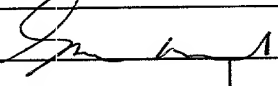
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DECLARATION

ADDITIONAL INVENTOR(S)
Supplemental Sheet
Page 3 of 3

Name of Additional Joint Inventor, if any:				<input type="checkbox"/> A petition has been filed for this unsigned inventor			
Given Name (first and middle [if any])				Family Name or Surname			
Michael Shane				O'Shea			
Inventor's Signature				Date		27/9/00	
Residence: City	Victoria	State		Country	AU	Citizenship	AU
Post Office Address 4 Worthington Court							
Post Office Address Mulgrave							
City	Victoria	State		ZIP	3170	Country	AU
Name of Additional Joint Inventor, if any:				<input type="checkbox"/> A petition has been filed for this unsigned inventor			
Given Name (first and middle [if any])				Family Name or Surname			
Graeme				Mead			
Inventor's Signature				Date		27/9/2000	
Residence: City	Victoria	State		Country	AU	Citizenship	AU
Post Office Address 9 Clarkmont Road							
Post Office Address Sassafras							
City	Victoria	State		ZIP	3787	Country	AU
Name of Additional Joint Inventor, if any:				<input type="checkbox"/> A petition has been filed for this unsigned inventor			
Given Name (first and middle [if any])				Family Name or Surname			
Inventor's Signature				Date			
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		ZIP		Country	

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